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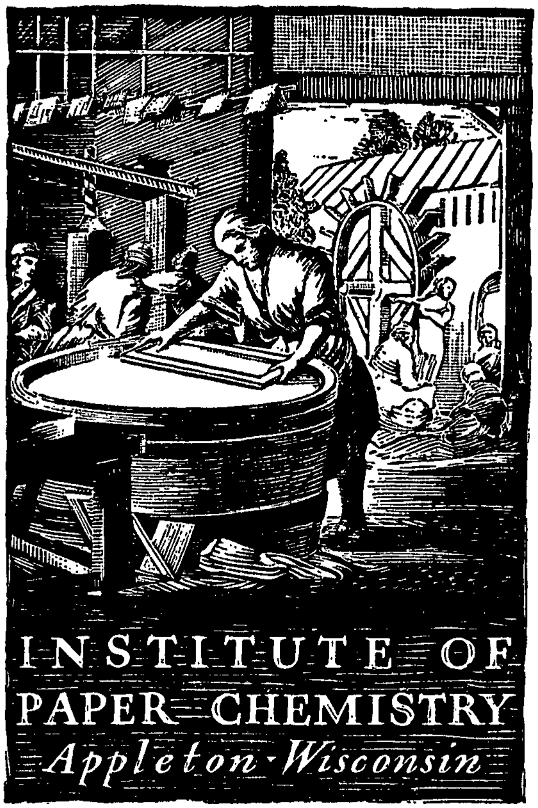

A STUDY OF THE EFFECT OF REPULPING ON FIBER PROPERTIES AND SHEET STRENGTHS

Project 1850-3

Report
to

JUTE RESEARCH GROUP

November 7, 1955

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THE INSTITUTE OF PAPER CHEMISTRY

Appleton, Wisconsin

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Progress Report One

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THE INSTITUTE OF PAPER CHEMISTRY

APPLETON, WISCONSIN

INTRODUCTION

It is well known that papermaking fibers undergo substantial changes in physiology when subjected to various environments common to stock preparation and machine processing such as beating, forming, drying, etc. For example, it is common knowledge that paper made from slush pulp will normally exhibit better strength properties than when made from the same pulp when in the dry lap form. Similarly, an air-dried sheet will exhibit somewhat greater strength than when dried at an elevated temperature. These examples illustrate in a small way the influence which the previous environmental history of the fiber may have on the behavior of the fiber upon being repulped and reprocessed. Investigations carried out in the field of waste paper regeneration have shown that the repulped or regenerated fibers (and papers made therefrom) differ significantly from the virgin fibers in many physical properties. The jute industry is undoubtedly beset by these phenomena as evidenced by the fact that it has not been feasible to develop the strength of the repulped fibers to the potential level exhibited by the fibers in the virgin state.

Since jute linerboard is manufactured primarily from waste paper, the nature and magnitude of the changes in the fibers, the mechanism through which they evolve and the extent of their influence on the properties of jute liner should be taken into account in any study related to the strength development of reclaimed fiber because these undoubtedly have an important bearing on stock preparation and subsequent machine processing. Consequently, a prerequisite to a general study of strength development is a knowledge of the effect which each phase of the stock preparation and paper machine processes has on the physical characteristics of virgin and repulped fibers and on the corresponding properties of the papers made therefrom. Once the nature and magnitude of the effect is known, attention can then be focused on determining the cause or causes responsible for these effects. Thus, the fundamental problem is one of developing basic data relative to the cause and effect as related to repulping of paper-making fibers. Once the fundamentals are known, studies of application of principles and equipment may be pursued more directly and efficiently in contrast to the costly and laborious trial and error method which must be resorted to in the absence of such information.

The Strength Development Task Committee of the Jute Research Group has inaugurated a study to develop this basic information and to apply the information in terms of principle and refining techniques.

The program as outlined is concerned with a study of the physical phenomena which influence the strength development of reclaimed fibers used in the manufacture of jute test liner. The program may be viewed as a four-phase study as follows:

Phase one is concerned with a historical review of available literature pertinent to the project. The purpose of this phase is to review the information available on the effects of stock preparation phenomena and machine processing environment on the properties of fibers and paper made therefrom.

The second phase is concerned with a repulping study. The purpose of this study is to develop basic information relative to the effect that the number of times a stock is repulped has on the morphology of the fibers, their physical properties and the physical characteristics of the paper made therefrom.

The third phase is concerned with analyzing the results of the experiment conducted in phase two. The purpose of this phase is directed toward the development of basic information regarding the mechanism responsible for the anticipated losses in strength properties. Thus, the combined efforts of phase two and three will provide information as to effect and cause.

The fourth phase is concerned with the application of basic information developed in phases two and three. The purpose of this phase is to study ways and means of developing a proportionately greater share of the initial strength of reclaimed fibers. This phase will also include new studies designed to clarify and elaborate on information obtainable from the experiment in phase two, but pertinent to an elucidation and understanding of the mechanism responsible for strength regression in repulping. This information will then be applied in an endeavor to propose a repulping technique which may counteract the adversities introduced as a result of the previous history of the fiber.

The study carried out to date has been concerned with phases one and two. The present report, therefore, covers the work completed relative to the historical review and the repulping study.

PART I. HISTORICAL SURVEY

According to the Dictionary of Paper (1) paper is defined in general terms as "the name of all kinds of matted or felted sheets of fiber (usually vegetable, but sometimes mineral, animal, or synthetic); formed on a fine wire screen from a water suspension. Paper derives its name from papyrus, a sheet made of pasting together thin sections of an Egyptian reed (Cyperus papyrus) and used in ancient times as a writing material." More specifically, paper made from wood or vegetable fibers may be considered as consisting of a mass of individual fibers meshed together with nearly all their longitudinal axes oriented parallel or nearly so to the surface of the sheet. In a handsheet the fibers are oriented randomly in the length-width plane but in a machine-made sheet there is a preferential alignment of the fiber in the machine direction. None of the fibers are straight, rather they are kinked, twisted and bent into an infinite variety of shapes. Even the fibers themselves are heterogeneous; they are composed of coarse and fine fibrils.

It may be deduced from the above that the strength of a sheet of paper is dependent on the strength of the individual fibers and/or the manner and tenacity with which the fibers are held together in a sheet, the latter being a function of the operations by which the fibers are prepared and formed into a sheet. Before elaborating on the general theory of paper formation and the development of the strength in a sheet of paper, it may be well to consider the physical structure of a wood fiber since that is the type of fiber we are concerned with in this study.

The shape of unbroken and unbeaten fibers or tracheids in kraft pulp made from coniferous woods--e.g., southern pine--may be likened to a long pencil-like configuration with rounded or pointed ends and a central cavity called the lumen. The shape of the cross section varies considerably although when viewed singularly in the wet state the cross-sectional shape tends to become circular. The length, "width" and wall thickness of fibers even from a given tree may vary widely; however, the average range of length is calculated to be in the neighborhood of 3-6 millimeters and the width 35-50 microns. On this basis it may be seen that the ratio of length to width is of the order of 100:1; thus, the thickness of the fiber is very small in terms of its length. According to the workers in the field of wood technology (2), each mature, woody fiber has a primary and secondary wall. The inmost boundary of the secondary wall defines the boundary of the lumen and the exterior surface of the fiber constitutes the primary wall. Mature cell walls have three major constituents--cellulose, hemicelluloses and lignin. The secondary wall which is the last part of the fiber to be formed by the protoplast varies greatly in thickness according to the nature of the fiber (e.g., the springwood in southern pine has a much thinner wall than summerwood). The cellulose structure in the fiber or cell wall can be broken down mechanically or chemically into fine threadlike structures known as fibrils, also referred to as windings. The fibrils can be seen with the aid of a compound microscope or can be made large enough for viewing by swelling in alkali. According to Brown, et al. (2) "they grade down in diameter from about one micron to the limit of visibility and probably far beyond." The alignment of these fibrils (the latter are believed to play an important role in papermaking) in the thin primary wall is not uniform and appears to approach a random orientation (3). Bailey and Kerr (4) have demonstrated the presence of three layers in the secondary wall. In terms of varying fibril orientation, these are a thin outer layer next to the primary wall, a central zone that varies greatly in width depending on the thickness of the secondary wall and a narrow inner tenuous layer bounding the cell lumen. As may be seen in Figures 1 and 2, in the outer and inner layers of the secondary wall, the fibrils are directed at right angles to the longitudinal axis of the fiber or form ascending counterclockwise helixes of very low slope. In contrast, the fibrils in the middle layer are oriented parallel to the longitudinal axis of the fiber or in counterclockwise helixes of steep slope. The exact role played by the fibrils in beating and forming has been a subject of wide interest. There is evidence that under certain bleaching and beating conditions the

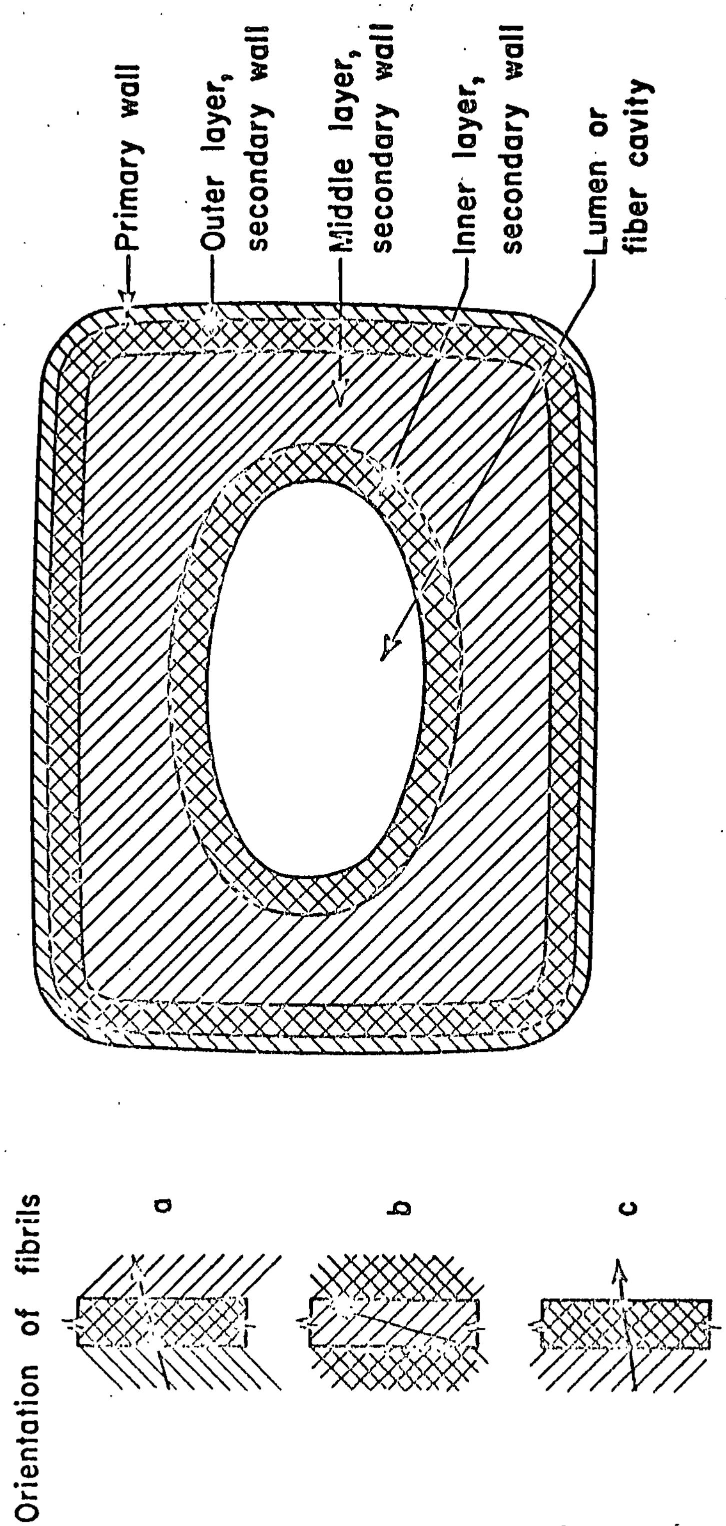

Fig. 1. Schematic Drawing of Cross-section of Fiber Showing Relative Position of Cell Walls

(a, b, and c, respectively, are the layers of the secondary wall, the arrows at the side indicate the orientation of fibrils. Longitudinal axis corresponds to the vertical.)

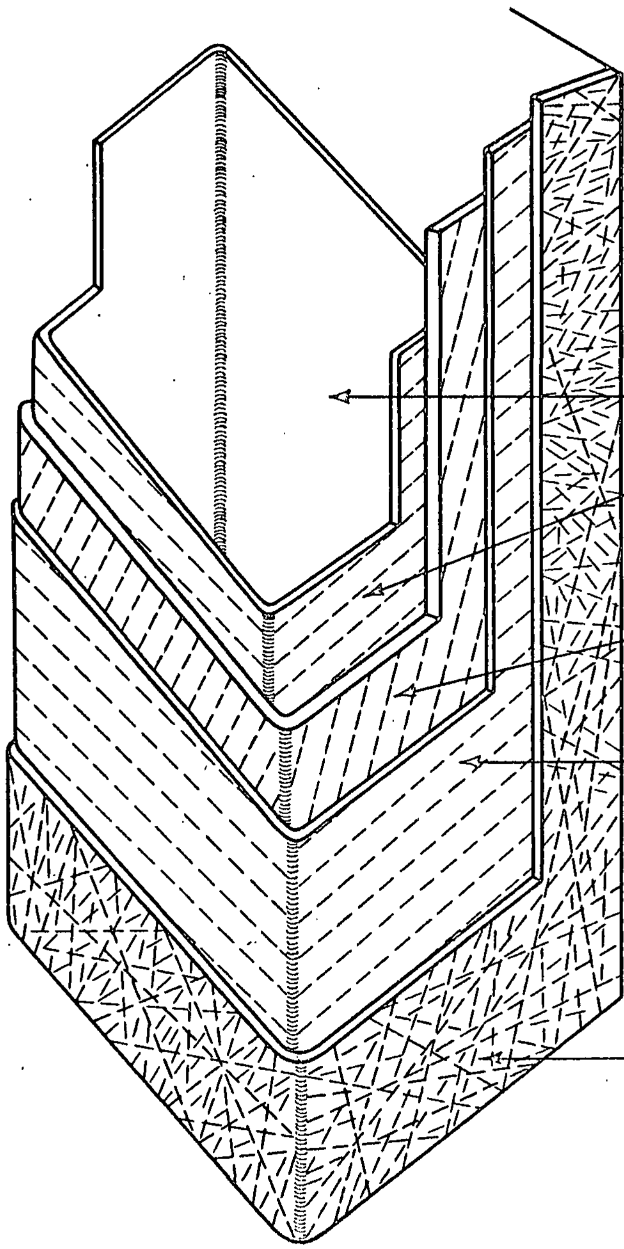

Fig. 2. Schematic Drawing of Fiber Showing Orientation of Fibrils in Fiber Walls

outer layers are ruptured at least in part and in swelling the fibrils unwind. This phenomenon is known as fibrillation.

The strength of papermaking fibers of the wood or vegetable type is developed usually by subjecting them to some mechanical attrition--beating or refining. The purpose of this action is to "open" up the structure of the fiber so that water can enter the amorphous cellulose portion of the fiber and swell it and also to condition the fiber as to devillication and length. The swelling appears to activate the surface of the fibers by creating at the fiber-water interface a partial "solution" of cellulose molecules which, according to present day theory, is the source of the bonding strength between plies. The swelling which takes place in conventional beating or refining is also accompanied by a cutting, bruising, crushing and fibrillation of the fibers. All these tend to reduce the intrinsic strength of the fiber, decrease the freeness and increase the resistance to drainage.

Early workers in the field of paper technology held to the theory that the strength of paper resided in the interweaving of fibers and fibrils. This concept has been discarded by most workers because paper made from highly fibrillated fibers and dried from a nonpolar medium such as benzene is extraordinarily weak. (<u>Note</u>: This procedure produces an unbonded sheet.) Also certain specific pulps which fibrillate extensively on beating will not produce a strong sheet when they are beaten and formed in water. The strongest argument against the mechanical binding theory is probably the relatively recent work involving stress-strain behavior. Paper exhibits both primary and secondary creep, the latter being an irreversible extension occurring on the first straining of the paper. Steenberg has shown that a strain-hardened paper will withstand repeated straining and relaxing without damage to its structure. This behavior would not be expected if the force holding the fibers together were primarily of a frictional nature. Also Steenberg shows that the load-elongation curves were in accord with the theory of Eyring and Halsey which does not embrace macroscopic slippage.

According to present theory, the fibers are held together by certain types of secondary valence bonds. When a fiber is beaten in water it swells and fibrillates, and the ends of the cellulose chain which are in the amorphous portion of the structure tend to protrude into the water layer. This action creates the effect of a saturated solution of cellulose over the amorphous portions of the fibers. As drying takes place, the surface tension forces compress the sheet and bring the fibers closer together. When fibers are brought so close to one another that these protruding portions of the fiber structure can intermingle, the latter will coalesce to form a bond when the water is removed from between them. Thus, paper appears to be a fibrous structure built up of intermeshed fibers held together by bonds of the secondary valence type. During drying the fiber surfaces are drawn very close together--within molecular dimensions needed for hydrogen bonding--according to the well-known theory of Campbell (5).

Because of the very small effective pore size of most papers and the distribution of fibers, the average fiber in a sheet is probably joined or bonded to many other fibers through these fiber-to-fiber bonds. This undoubtedly takes place to an even greater extent in the case of a handsheet because of the random distribution of the fibers in the plane parallel to the normal surface. The failure of a sheet of paper results from a combination of (a) rupture of the individual fibers and (b) a pulling apart of the fibers because the resistance of the bonded area stressed is exceeded. The resistance of the bonded areas is dependent on the strength of each bond, and the number of bonds. The breaking length of individual pulp fibers has been given as being in the range of 40,000 meters and the breaking length of paper in the range of 5000-9000 meters. Part of the difference is due to the random orientation of the fibers in the sheet; however, it would appear on the basis of this difference that breaking length is primarily a function of the number and strength of the fiber-to-fiber bonds.

A great deal of time and effort has been devoted to investigations directed toward determining what happens in beating and refining. One of the early theories sponsored by such early investigators as Cross, Bevan and Schwalbe (6, 7) was the chemical theory.

This theory suggested that cellulose formed a hydrate in water; however, the theory has been abandoned because it has been proved that cellulose does not combine with water to form a hydrate. Further, it is believed that no "major" chemical change takes place during beating, as evidenced by the x-ray diffraction pattern which does not change on beating. Minor chemical change may occur as a result of the great increase in surface activity of the fiber and other changes in the physical properties of the fibers (8, 9).

In 1926, Strachan (10) attacked the chemical theory of beating and suggested the physical theory of fibrillation. According to Strachan's ideas, the so-called hydration obtained on beating is really fibrillation of the fiber surface which increases the specific surface and the amount of water imbibed. Strachan (11) originally visualized the beating process as the development of a fine "pile" of fibrillae on the surface of the fiber somewhat analogous to the pile on velvet with the effects being largely confined to the surface of the fiber where they produce a tremendous increase in the external specific surface. The presence of this fine fuzzlike material accounts for the tremendously large surface of beaten fibers. Thus, he visualized beating as producing an increase in the external surface of the fibers at the expense of the internal surface, but not affecting the total surface. Strachan (12) later observed the presence of fibrils when examining the surface of relatively unbeaten fiber with the microscope and assumed that they were bundles of micro-fibrils about 0.2 microns in diameter. Sears (13) also observed the presence of microfibrils and noted still finer fibrils which he estimated to be 50 millimicrons (500 A) or less in width. Others like Clark (14) indicate that these fine fibrils may be covered with even a finer fuzz of molecular dimensions. Clark also believes that the fine fuzz is cemented to the body of the fiber on drying and does not reappear on wetting, unless the pulp is again beaten.

The phenomenon of beating by the fibrillation theory, as presented by Strachan (11, 12), may be viewed as follows: Initially, the fibers imbibe water and swell, which results in a weakening of the cohesive forces. In this wet swollen condition, the fibers are low in strength and very soft and plastic so that the mechanical action of the beater tends to loosen the individual fibrils. At the same time, still finer fuzzlike material which may consist of individual cellulose molecules or fine crystallites, is raised on the surface of the fibrils. The loosening of fibrils within the fibers (internal fibrillation) increases the amount of surface available for contact. These two actions tremendously increase the external specific surface of the fiber and the amount of water which is retained in the fiber network.

Strachan (15, 16) has also presented an electronic theory to explain fibrillation in which the displacement of each fibril on the surface of the fiber requires a definite expenditure of energy, and the amount of this energy is dependent on the magnitude of the force holding the cellulose crystallites together.

In 1932 Campbell (17) proposed the partial solubility theory. This theory is based upon an earlier hypothesis of Urquhart (18), which suggested that cellulose is soluble in water in certain stages. The solubility theory is based upon the supposition that the cellulose crystallites on the surface of the fiber becomes partially "dissolved" in water and in this state attach themselves to similar crystallites on adjacent fibers. While in this state of near solution, the molecules or crystallites are endowed with a certain freedom and can arrange themselves so that they are pulled together by secondary valence forces when the water is removed. In this way, the fibers are united by the crystallization of the cellulose during the evaporation of water.

Since the original proposals of these three mentioned theories--namely, the chemical theory, the physical theory, and the partial solubility theory--there have been other modifications and variations which are worth mention.

Clark's theory (19) is somewhat similar to that of Campbell and it is said that the two theories, taken together, furnish a mental picture of what is meant by "papermakers hydration." Clark regards the surface of well-beaten fibers as a "two-dimensional colloidal system" in which the surface fibrillae have two dimensions in the colloidal range, but are anchored to the fiber in the third. Thus, these fibrillae, which are made up of units at least as thick as cellulose crystallites act almost as if they were in colloidal solution. The degree of adhesiveness between fibrillae thus depends on the degree of colloidal solubility, which is inversely proportional to the degrees of polymerization of the cellulosic and hemicellulosic material, making up the fibrillae. Combining Clark's and Campbell's theories then, the surface of the fibers may be visualized as covered by water molecules which are so strongly adsorbed and oriented that they "hydrate" the surface.

Edge (20) indicates that beating accomplishes another important function, namely, the freeing of hemicellulose. He holds that in the process of beating, the primary cell wall wrappings are broken, freeing the hemicelluloses from the interior of the fiber; these then become adsorbed on the externally exposed fiber surfaces and aid greatly in the effectiveness of the surface tension forces that draw the structure together, and they serve also as a gap-filling glue to increase the extent of fiber-to-fiber bonds.

Investigators in the field of stock preparation and paper strength seem to agree that the degree of swelling or the amount of water imbibed or held by the fibers is an important consideration in relation to pulp and stock preparation, sheet formation, bonding and the properties of the finished sheet. The manufacture of paper involves a swelling of the fibers during the early stages of the general process and a reversal or deswelling during the drying of the sheet. It is of importance, therefore, to consider the nature of the deswelling, the nature of the factors which are known to bring about the increase in swelling and its relationship to paper manufacture and strength properties. In the case of jute linerboard, all the pulps and papers used in producing the linerboard have been previously dried and the effect of this drying undoubtedly has an important bearing on the use of these materials.

Heath and Johnson in a discussion of their work relative to the proportion of water-accessible and nonaccessible cellulose in wood pulp (21) state "that the structure of a cellulose system is a mixture of two solid stages of aggregation which are interconnected because individual cellulose molecules may extend from one state to the other and thus exist in both states simultaneously." The two states are the crystalline, which produces an x-ray diffraction pattern similar to a crystal lattice, and the amorphous, which produces only a diffuse background in the x-ray diagram. The crystalline areas have the characteristic properties of crystals--that is, greater density and tenacity but low elasticity and flexibility; they are nonreactive and difficult to swell or penetrate with liquids. The amorphous areas, although they show wide variations from point to point, should be more porous, more flexible and more readily swollen.

According to Sisson (22) the swelling of cellulose in water is essentially intermicellar--i.e., the water is imbibed or held for the most part in the amorphous regions of the fiber without alterations of spacings in the crystalline region. Young and Rowland (23) have shown that the hemicelluloses are of particular importance in determining the degree of swelling of the fiber. Cellulose is hydroplastic and not thermoplastic in contrast to most linear polymers and, therefore, the degree of swelling cannot be enhanced by heat (24). It is well known that the drying of pulp brings about changes in the properties of the fibers which do not revert wholly to the original condition even after prolonged soaking in water. The effect of drying on the degree of swelling of the fiber is very marked.

Jayme (25) has conducted extensive swelling tests on fibers to demonstrate the effect of wetting and drying. In this respect, Jayme noted that cellulose fibers when dried lose much of their ability to swell to their original swelling value. The inability of a fiber to return to its predrying swelling value, is termed by Jayme as "irreversible cornification." According to Jayme, this loss in swelling potential is not fully recovered even after prolonged soaking. Jayme also noted that consecutive wetting and drying causes a decrease in specific volume and effective specific surface. Jayme and his co-workers (26) also conducted swelling experiments in various concentrations of sodium hydroxide. The maximum swelling value was of the order of 350% and the maximum swelling occurs at a concentration of 10.5% by weight of sodium hydroxide. In the absence of a hydrate the maximum was attained with 3% sodium hydroxide.

Mason (27) has noted, from reviewing studies concerned with measuring the specific external surface area of pulp, that a correlation is believed to exist between the effective specific volume and the development of strength by beating. From these and other results, Mason states that one is tempted to conclude that the real action of the beater in developing strength is not in producing fibrillation and increasing specific surface, but rather in inducing swelling in the fibers by the mechanical action of the beater. Mason goes on to say that, "it is possible that swelling, accompanied by a softening and flexibilization of the fiber is what the practical papermaker means by the term, 'hydration,' and not the increase in specific surface, as measured in freeness and drainage tests." Mason, however, indicates that this opinion should be treated with reserve, since the factors involved in high specific surface values are still in question. The combined results of Robertson and Mason (28) further indicate that there is a relationship between fiber swelling during beating and the strength properties of sheets made from the swollen pulp. The relationship is illustrated by plotting bursting strength and effective specific volume against specific surfaces for fast beating and slow beating—i.e., beaten to the same freeness level with varying rates of beating. In each case the bursting strength increases as the effective specific volume increases. The increase in effective specific volume is in turn shown to be dependent on the swelling power of the swelling medium.

Since the swelling of cellulose is an exothermic reaction, the degree of swelling will decrease with temperature. Lewis and Gilbertson (29) in a study of the effect of temperature on the beating of rag stock found that within the temperature range of approximately 34 to 185°F. the rate of beating, as measured by time to a given freeness, was lowest at 185°F. and highest at 77°F. Starting at 34°F., the rate of beating appeared to increase to a maximum at 77°F. and then decrease to a minimum at 185°F. The physical properties such as fold endurance, bursting strength, internal tearing strength, and tensile strength were all lower at the higher temperatures. Noll (30) beat wood pulps at various temperatures and found that regardless of type of pulp, a retardation of the beating effect took place at rising temperatures including all physical constants such as freeness, tearing strength, bursting strength, etc. He suggests that since a longer period is required for obtaining the same degree of beating at higher temperature, power consumption is naturally greater. He further states that the chemical constants do not influence the thermal behavior of a pulp whereas the swelling properties are in direct relationship to the different temperatures. Noll interpreted this retardation as a function of the lower surface tension of the water at higher temperature.

Heath and Johnson (21) mechanically separated Douglas-fir pulpwood into its springwood and summerwood constituents and each was separately pulped with the same kraft cooking procedure. The water accessibility characteristics were determined on the pulps as received; another set was made on handsheets dried in the usual way against a steam-heated metal surface, and a third set of handsheets was allowed to dry at room temperature. In all cases springwood was more accessible to water than summerwood. Further, the air-dried handsheets of both springwood and summerwood exhibited considerably higher accessibility than the corresponding sheets dried at an elevated temperature. This suggests that secondary valence bonds may be formed in the amorphous portion of the cellulose on drying which decreases the accessibility of the fiber to water.

Although it is well known in the industry that the drying of pulp reduces the paper strength, it would appear that relatively few data have been published giving quantitative effects. The effect of drying of pulp on the bursting strength of paper is very marked. Data secured in a survey of mill pulp drying (31) showed large losses ranging up to 73% of the original wet pulp strength. Lyne and Galley (24) found that there appears to be a linear relation between loss in bursting strength and the dryness of the pulp from which the sheets were made. The mill survey mentioned above (31) showed that the progressive loss in bursting strength with increased pulp dryness is much less on a beaten pulp. It is apparent that the effect of the beating following partial drying assists greatly in the reversion of the pulp to a state of greater swelling ability. Babbitt (32) has shown that vacuum-dried pulp possesses higher strength than air-dried pulps when both are beaten and a similar conclusion was reached by Kehoe (33). Minton (34) states that "it is well established that low temperature will not hurt the papermaking fibers and that high temperature will injure them. It has also been pretty well proven that when papermaking fibers are wet they can be subjected to quite a high temperature without injury." It seems to be held generally that the lower the drying temperature the stronger the fibers will be in the dry state. The investigation carried out by Minton showed that when a given furnish is dried at 100°F. (using vacuum) the resulting product will be much stronger than when dried at a higher temperature. Minton suggests that the loss in strength may be associated with "oxidation." He further states that his studies show that the drying temperature affected fourteen different properties among which were bursting strength, tensile strength, internal tearing resistance, fold endurance, and stretch. Presumably, the strength characteristics decreased with increasing temperature although no direct mention is made or data presented.

Lyne and Galley (24) found that the effect of drying pulp on tensile strength and fold endurance was similar to that noted for bursting strength. In the case of tearing strength, it increases progressively up to about 60-70% solids, following which it decreased sharply up to 100% solids. The large decrease in tear in thoroughly dried pulps results presumably from an embrittlement of the fiber and an excessive slippage resulting from lack of bonding.

Stamm (35) has expressed the view that shrinkage of the fiber and bonding on drying set in as the imbibed water is removed. According to Lyne and Galley (24), drying is accompanied by a shrinkage of each fiber. Since bonding sets in during drying, the relationship between the extent of shrinkage and the water content of the web is of importance. They measured the shrinkage force during drying and found that shrinkage developed rapidly at about 55% solids and was apparently complete by 80% solids. Tensile measurements during drying were also carried out which showed that strength development begins at about 55% solids and increases rapidly to 80% solids. From the results, it appears that bonding and shrinkage both commence at the same stage of dryness. This is where the free water has been essentially removed and water of swelling begins to diffuse to the surface and the degree of swelling undergoes a change--deswelling.

Casey (36) states that cellulose has a high molecular cohesion but if the penetrating agent is strongly polar in nature, swelling of the fibers results. There are two types of swelling, namely, swelling of the crystallites and swelling of the intercyrstalline, depending on whether the swelling agent penetrates into the crystalline or only into the intercrystalline region. Cellulose exhibiting the highest percentage of amorphous material exhibits the greatest amount of swelling since water preferentially enters the amorphous region.

Casey indicates that cellulose will also undergo considerable swelling in the presence of certain salts and acids, and may actually dissolve in these substances if the concentration is high enough. The extent of swelling in salts depends upon the hydration of the ion, and consequently, the salts which "dissolve" cellulose are those ions whose molecules are highly hydrated. Strong solutions of mineral acids (hydrochloric acid, 40%, sulfuric acid, 70%) swell and will also dissolve cellulose.

Weidner (37) noted that cellulose fibers swell appreciably in width but very little in length. This difference in expansion in the lengthwise and crosswise direction he attributes to the highly organized nature of cellulose fibers resulting from the orientation of the crystallites and fibrils in predominantly the same direction as the fiber axes. Thus, since water penetrates fairly readily between the crystallites (and only with difficulty into the crystallites), it pushes these units apart in a lateral direction, but causes practically no longitudinal expansion.

According to Urquhart (18), the ability of a pulp fiber to swell is attributable to their hydroxyl groups, since without these polar hydroxyl groups, cellulose fibers will not swell in polar liquids. Kress and Bialkowsky

(8) measured the swelling of cellulose fibers in various liquids and found that highly polar liquids such as water, formamides, and ethylene glycol result in considerable swelling, whereas nonpolar liquids result in very little, if any, swelling. They found that water and ethylene glycol resulted in a 90% increase in volume, and formamide in a 155% increase in volume, whereas n-propyl alcohol results in only a 6% increase. Only those liquids which show a high swelling tendency toward cellulose permit separation of the fibrils on beating.

Morgan and Libby (38) noted that pH was an important variable in beating cotton pulps. Their results indicate that minimum strength in beating was obtained at a pH between 4.0 and 4.5 and at a pH of 10. The maximum strength on beating was obtained at a pH of 8.5 which is close to optimum value--i.e., pH 8.0, reported by Renaud (39). Hansen (40) also found that the beating of wood pulps was retarded by low pH values and was speeded up in alkaline mediums in accordance with the greater swelling of cellulose fibers in alkaline medium. Other investigators have noted a minimum in both the swelling and degree of beating at a pH around 7.0 to 7.5.

Campbell (5) has shown how fibrillation leads to improved formation and densification of the sheet. The high affinity of the fibers for water that results in a higher interfacial surface tension between fibers and water than the high surface tension of water itself, thus causing the water draining through a sheet being formed on the wire to pull the fibers down to the wire. As drying progresses and water is no longer in a continuous phase but in films between fibers, surface tension forces continue to act with increased effectiveness. Two parallel fibers with a film of water with concave meniscuses between them are pulled together with a force equal to twice the surface tension of water. This drawing together will continue until it is balanced by the structural resistance. The structural restraint to bringing of irregular surfaces together will be much lower for the most flexible fibrils than for the fibers. Then, too, the surface-tension force that is the same between fibers of any size is far greater between fibrils than between whole fibers per unit of cross section. Campbell (5) has computed what these forces would be under different conditions. For example, the attractive force between fibers 3 by 10^{-3} cm. in diameter is 87 p.s.i.; that between fibrils 2 by 10^{-4} cm. in diameter is 540 p.s.i.; and that between fibrils 2 by 10^{-5} cm. in diameter is 2480 p.s.i. This high internal tension between fibrils far exceeds the normal externally applied compression in the paper machine. It is for this reason that a beaten pulp shrinks considerably more during the papermaking process than an unbeaten pulp and results in a denser sheet. Formation of sheets from unbeaten pulp under the high pressure so as to attain the density of a sheet made from beater stock has been shown to produce practically as strong a sheet. Fibrillation in a beater can thus, in part, serve as a substitute for applied pressure in sheet formation.

The effect of formation on the paper machine wire influencing sheet strength is indicated by the results of Cottral and Gartshore (41). Cottrall and Gartshore found that inferior make on the wet end of a Fourdrinier machine results in a loss of strength of the order of 25%.

Mason (42) indicates that some of the most important factors influencing sheet formation are fiber length, shape, consistency, and the conditions of motion to which the suspension is subjected.

The drainage resistance of a survey of fibers in water has an important bearing on the papermaking characteristics of the stock because of its influence on flocculation, speed and formation. Ingmanson (43) has shown that the Kozeny-Carman equation, which relates the rate of flow of an incompressible fluid through a fixed porous bed to the specific surface, the effective specific volume and the compressibility of the bed, may be applied in differential form to a mat of pulp fibers and then integrated over the proper boundary conditions. By this means, the filtration resistance can be expressed in terms of its components. Ingmanson and Whitney (44) believe that the filtration resistance is an important property of a pulp slurry and may be related to the drainage of the stock on the wire or cylinder. "When measured on a whole pulp at a slurry consistency free from flocculation effects, filtration resistance may be defined as the maximum possible value to be expected on the machine wire. The related effects of pulp flocculation and loss of fines experienced in actual machine operation will result in lower filtration resistance +++." The filtration resistance provides only information relative to drainage behavior. It does not tell anything as regards the potential strength properties. The components of the filtration resistance--specific surface, the effective specific volume and compressibility, however, are related to strength potentials. Knowledge of the influence of these physical properties on ultimate sheet strength may assist in the selection of refining conditions that represent a maximum development of strength for certain desired levels of filtration resistance.

As was mentioned previously, the "beating" action tends to reduce not only the intrinsic strength of the fiber as a result of crushing, mashing, kinking and the unwinding of the fibrils (fibrillation) but also there is a marked tendency towards reducing the length of the fibers by cutting or shearing. The influence of the length of the individual fibers on the strength characteristics of paper is still a controversial subject. It is generally agreed that fiber length exerts its greatest influence on the internal tearing strength. Theoretically, it would seem that the greater the fiber length, the greater would be the ultimate strength of the paper since each fiber would be bonded to a larger number of adjacent fibers by a larger number of fiber-to-fiber bonds. These appear to be, however, an optimum fiber length for good sheet characteristics. If the fibers are too long, sheet formation is poor. As pointed out by Van den Akker (45), the average fiber in a sheet of paper is bonded to so many other fibers that the intrinsic bonding strength does not need to be exceptionally high for most ordinary papers. On this basis, it might be argued that the fibers would fail before the rupturing of bonds in a hypothetical paper containing very long fibers on the ground that each fiber is so well bonded to other fibers. As he points out, failure initiates in a zone where the local stress and strain are high. In such a zone, the fibers become elongated beyond the average value, and bonds in and near the zone become involved in a stress concentration that could cause them to rupture irrespective of what length of fiber and what number of bonds extended beyond the zone.

The effect of the distribution of the fiber length on the strength of paper has not been considered extensively. No conclusions have been reached as to whether or not it would be advantageous to have a comparatively homogenous distribution or a completely heterogeneous distribution of fiber length. In general, it is considered that the longer the fiber length, the poorer the formation because of the greater tendency for the fibers to flocculate, other things equal. In recent years, there has been some controversy regarding the influence of fines on the strength of paper. Keeney (46) found that an increase in fines produced an increase in tensile strength. He postulates that the most likely manner by which the fines could increase the tensile strength is through an increase in the number or strength of the fiber-to-fiber bonds; the fines act as cross linkages between the fibers.

According to Lathrop and Naffziger (47), the relationship which exists between long and short fibers is illustrated in fiberboards which normally consist of different ratios of long and short fiber fractions. The long fibers act as beams or girders in the board and are principally responsible for the structural or impact strength of the board. On the other hand, the short hydrated fibers act as the cementing agent and consequently tend to increase the density and the flexural and tensile strength (47, 48).

Fiber length is an important index of fiber quality, but it is subordinate to the swelling and fibrillating qualities, since adequate sheet strength can be obtained only by means of a high degree of fiber-to-fiber bonding, which prevents the fibers from slipping past one another. This is indicated by the results of Jayme (49) who found that the bursting strength of straw pulps, which have an average fiber length between 0.30 to 0.46 mm., may approach under special conditions the bursting strength of spruce pulps having a comparable average fiber length between 1.2 and 1.8 mmu.

As may be seen from the foregoing, most of the investigations carried out relative to pulp and stock preparations, forming, drying, and sheet properties, have been directed to the environments which might befall virgin stocks. In the study at hand, we are concerned with the mechanisms involved in repulping papers. Thus, all the foregoing applies in part or whole to each and every cycle of repulping to which the papers are subjected. The extent of the influence of "beating," forming, drying, and finishing on the intrinsic fiber properties, the filtration resistance, the degree of swelling, and the strength and number of fiber-to-fiber bonds may vary, however, with each repulping cycle. The literature on the subject of repulping is almost nonexistent. A few studies have been carried out. However, most of these were unfortunately directed only to determining the extent of the affect of repulping on strength properties and apparently no attempt was made to try to relate the change in sheet properties to fiber or stock properties. Pfaler (50), using a furnish corresponding to newsprint, found that, on repulping, the bursting strength decreased about 16%, tensile strength 10%, and tearing strength approximately 6%. They stated that the groundwood fraction apparently underwent less degradation than the sulfite fraction.

Brecht (51) carried out an extensive study to determine the effect of repulping on a furnish consisting of northern unbleached sulfate pulp without the addition of size or filler. Care was taken that the furnish conditions of sheet making and disintegration was always identical. The results showed that for equivalent beating time the repulped stock was always freer and the sheets made therewith less dense and more absorptive than those prepared from the same stock initially. Tensile strength, fold endurance and internal tearing strength were usually considerably below the original values, the only exception being very "slow" papers. Brecht attributed these changes to loss of fines. Brecht found that the essential properties of the original sheet could be maintained through several repulpings when a slight amount of fines was added each time to the stock. Wet pressing of repulped stock increased the density and many strength factors to a certain extent; however, it did not equal the results obtained originally. Thus, the results show that the use of high wet pressing does not replace in full the role played by the fines. As pointed out previously, the fines undoubtedly help in the bonding.

A cursory study relative to the effect of repulping on the physical characteristics of paper made thereby has been carried out by Shartles Bros. Machine Division of Black-Clawson (52). The results obtained on repulping old kraft corrugated stock indicated a rise in freeness in repulping. Although no mention is made in the report as to the exact procedure used, it appears that the furnishes were all subjected to the same beating conditions, particularly that of time. This is, however, a rather unrealistic approach. On the basis of the approach used, the results indicate that the bursting strength, tensile, tear, and fold endurance all decrease with the number of times repulped. The order of magnitude of these strength reductions are given as 66, 18, 33 and several hundred per cent for bursting strength, tensile, tear and fold, respectively. Trials were also made to determine the effect of adding new kraft corrugated stock to stock which had previously been repulped 1 to 4 times. It is believed that the new kraft corrugated was refined separately and then added to equal portions of stock repulped 1 to 4 times. In discussing these results, the author states that "a very considerable percentage of repulped stock can be used without seriously affecting the final strength of the product being manufactured." Another trial using 100% unbleached kraft (presumably pulp, which had been refined to a freeness of 335 cc. Canadian Standard was used). This stock was dried, repulped, dried and repulped and tested. The results of one initial and two repulpings show that bursting strength, tensile and fold decrease with an increase in freeness and tearing strength.

On the basis of the foregoing, it may be seen that, in the process of making pulp into paper, many physical and chemical phenomena are brought into play which have a profound influence on the fibers. The first major operation is that of preparing the fibers by beating or refining so that they will take up water and swell, will become crushed, kinked, twisted, cut and in a sense unravelled. The next major operation is that of forming the thus prepared fibers into a uniform mesh or web in such a manner that the inherent strength potentials are fully realized in the final sheet. The third major operation is that of drying which takes place under tension and at an elevated temperature. It may seem paradoxical that, in drying, we, in a limited sense, destroy many of those fiber properties which required so much energy to develop earlier--particularly the degree of swelling. On the basis of swelling alone a great deal of energy is expended in getting the fiber to swell and then more must be expended in deswelling. Each of these have a marked influence on the properties of the fibers. Thus, it is not surprising that paper made from repulped stock will exhibit a strength depreciation since no steps are employed in the repulping operation to correct for the physio-chemical changes induced in the fibers by the environments to which it was exposed in the initial processing into paper.

PART II. REPULPING STUDY

A. INTRODUCTION:

As previously mentioned, the second phase of this study was concerned with carrying out a series of repulping experiments for the purpose of determining the effect which progressive repulping has on the properties of the fibers and the sheets made therewith. For this purpose, unbleached virgin kraft pulp was used as the initial raw material. There are a number of other fibrous materials which could have been used; however, the virgin kraft was selected because a very high percentage of corrugated board, from which come the bulk of our waste fiber for jute linerboard, is now made with kraft liners. Further, analysis of the fibrous content of jute linerboard shows a high kraft fiber content.

The general approach used in this phase was to defiber and refine the virgin pulp to a freeness of 615 $\pm$ 15 cc. (Schopper-Riegler) in a laboratory Valley beater. After beating to this level, all the stock except that used for stock tests was made up into handsheets, dried on a steam-heated drier to simulate as nearly as possible machine conditions, conditioned, and tested. The stock test made included pH, freeness, degree of swelling, fiber length classification and filtration resistance. The tests performed on the handsheets were as follows:

1. Basis weight
2. Caliper
3. Apparent density
4. Bursting strength
5. Tensile
6. Bonding (transverse)
7. Stretch
8. Elmendorf tearing strength
9. Taber stiffness
10. Fold endurance
11. Porosity
12. Bonded area
13. Zero span tensile

The heat-dried handsheets from the first beating cycle, corresponding to zero repulping, which were not used for test purposes became the raw material for the second beating cycle. This latter corresponded to the first repulping cycle. This procedure was used for the initial refining and for the six subsequent repulping cycles, each time the stock was refined under the same conditions to the same freeness level, namely, 615 cc.

Since the stock tests, test sheets, etc., necessitated the removal of a considerable quantity of fibrous material from each beater run, it was necessary to use an initial quantity of pulp equivalent to about twelve pounds on an ovendry basis in order to have sufficient material for all the repulping cycles. Inasmuch as each beater run was charged with the equivalent of 360 g. (ovendry) pulp it was necessary to make fourteen beater runs in the initial cycle. Taking into account the stock used in the stock tests and the losses due to dumping the beater, the average fiber yield per beater for making up into handsheets amounted to about 265 g. This quantity of stock made about 140 handsheets, 8-inches in diameter at 45-lb. ream (25x40--500). Because of these unavoidable fiber losses it was necessary to decrease the number of beater runs at each succeeding cycle. A total of seven beating cycles (initial and six repulpings) was used. The number of beater runs at each cycle is shown in Table I.

TABLE I

NUMBER BEATER RUNS AT EACH CYCLE

Cycle	Number Times Pulped Previously	Number Beater Runs Made
0	0	14
1	1	10
2	2	8
3	3	6
4	4	4
5	5	3
6	6	2

B. MATERIAL USED

As previously mentioned, the starting raw material consisted of virgin unbleached kraft pulp. The pulp was obtained from Container Corporation of America and is marketed under the name of Conus Pulp. This pulp is a normal yield, southern kraft pulp and was received at The Institute of Paper Chemistry in baled sheets.

C. GENERAL PROCEDURE

1. Preparation of Pulp

When the pulp was received at the Institute, the sheets were cut into six-inch squares and rotated in a 7-foot revolving drum for six hours over a two-day period in an atmosphere maintained at 50 ± 2% relative humidity at a temperature of 73 ± 3.5°F. to permit equalization of moisture content and to afford good mixing. After the prescribed period, the twelve-pound sample used in the initial beater run was removed. After removal from the drum the 12-pound sample was further conditioned for four days and then checked for moisture content. An amount of pulp equivalent to 360 ± 1 g. of ovendry fiber was weighed out for each of the fourteen beater runs carried out in Cycle O. The individual beater charges were stored in polyethylene bags until they were used. Prior to beating, the pulp was soaked in water at 73°F. for four hours prior to being charged to the beater.

2. Beating Procedure

Institute Method 403-3 was employed in beating the pulp to a freeness of 615 ± 15 cc. Schopper-Riegler. This corresponds to a Canadian Standard freeness of approximately 320 cc. Slight deviation from the Institute Method (403-3) are noted as follows:

a. The soaked pulp was not defibered in the disintegrator prior to beating in the Valley beater.

b. Slushing of the pulp was performed without a balance weight on the bedplate level arm. The beating was carried out using 5500 grams plus the balance weight on the bedplate arm.

3. Stock Sampling and Testing

a. Freeness. Five hundred milliliter samples were removed from each beater run after the five-minute slushing period as well as periodically throughout the beating cycle for the purpose of checking the freeness. These five hundred milliliter samples were diluted to 1500 cc. (0.523%) and a 250-ml. portion was used to check the consistency in the beater. The remainder was used for duplicate freeness determinations. Institute Method 414 was used to determine Schopper-Riegler freeness.

b. pH. Samples for pH determination were removed after five minutes slushing and at the end of the beating cycle. The pH was determined using a Beckman pH meter.

c. Fiber Length Classification. Bauer-McNett fiber classification determinations were made on two beater runs in each cycle. For this purpose a thirteen hundred milliliter sample was withdrawn at the end of the designated beater run. As an aid in characterizing the original pulp, fiber classification tests were made on the stock in the first cycle after five minutes of slushing. The screens used in the Bauer-McNett were 20, 35, 65, and 150.

d. Degree of Swelling. The degree of swelling was determined on the stock prepared in each repulping cycle. For each test a one hundred milliliter sample was removed from the beater after five minutes slushing and also at the end of the beating cycle. In order to minimize the influence of a given beater, swelling tests were made on every odd numbered beater run in a given repulping cycle. Thus, on the initial cycle, swelling tests were made on the 1, 3, 5, 7, 9, 11 and 13 beater runs. The procedure used in determining the degree of swelling was a centrifuge method similar to the one employed by Jayme (49, 53). The essential steps were:

1. Weigh a quantity of pulp slurry into a (Jayme type) centrifuge bottle. (See Figure 3)
2. Centrifuge at a given r.p.m. for a specified time.
3. Weigh the pad after centrifuging
4. Dry pad at 105°C. to constant weight.

The swelling is calculated in per cent from the following relations:

$$\frac{\text{Moist pad weight - dry pad weight}}{\text{Dry pad weight}} \times 100 = \text{degree of swelling}$$

The use of this method for determining the degree of swelling is based on the concept that, when a fiber swells, it takes up water called imbibed water. This water is held rather tightly by the

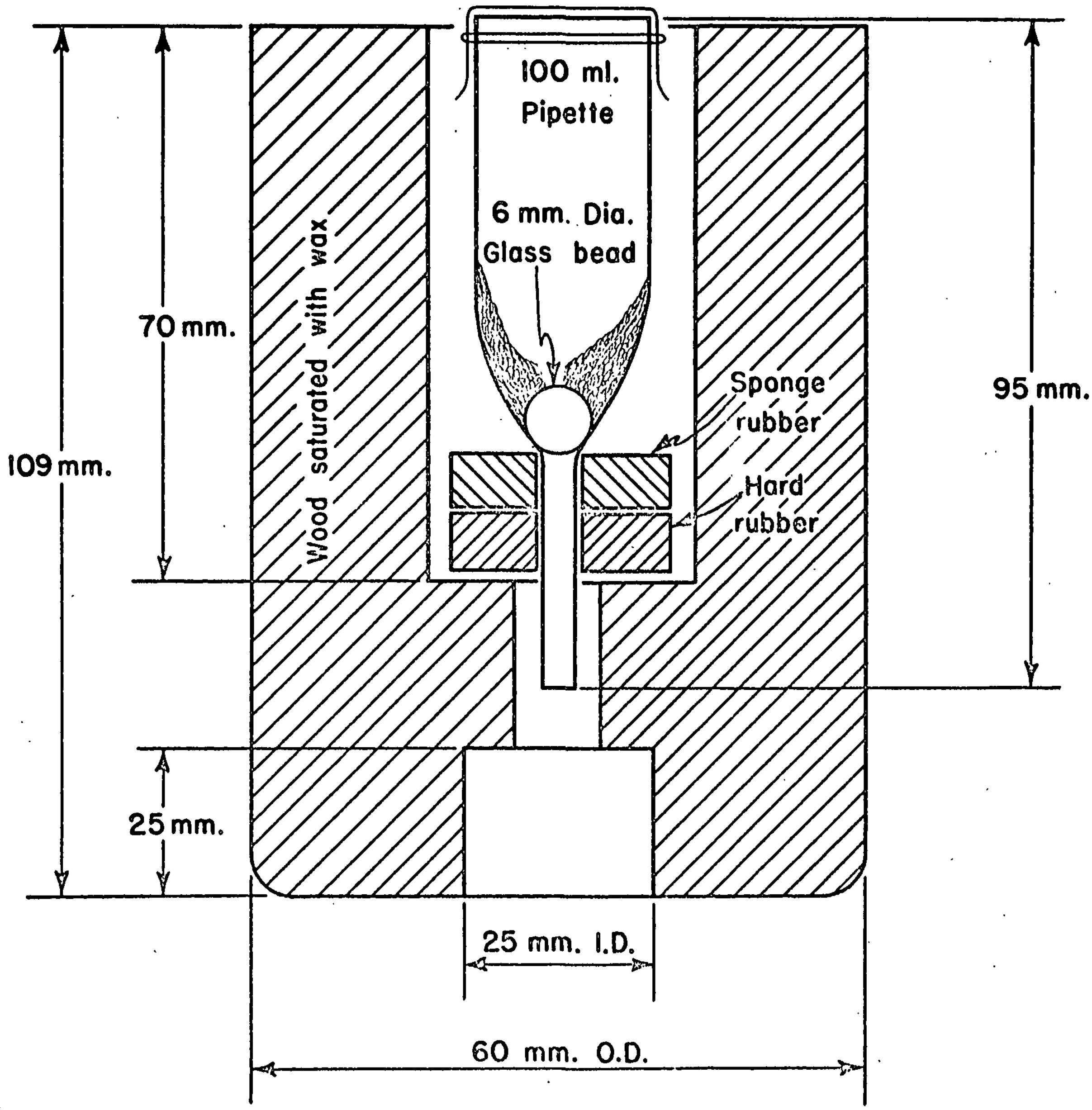

Fig. 3. Centrifuge Cup, Glass Bead and Holder

fiber. Further, the degree of swelling is proportional to the amount of imbibed water. On centrifuging, the free water--i.e., that not held by the fiber--will be removed easily but the imbibed water is held too tightly to be removed in the centrifuge. Although this method is open to considerable criticism, it does serve as a quick and reasonably reliable method, particularly when used for obtaining comparative data.

The apparatus used for the above-mentioned swelling tests was an International Centrifugre fitted with large-type centrifuge cups. Inserts for the centrifuge cups were fabricated from wood, saturated with wax to prevent the absorption of moisture. These wooden inserts thus served as a container for holding the "Jayme Type" centrifuge bottles. Since the centrifuge bottles were fabricated from 100-ml. pipettes, they were delicate and required sponge rubber inserts to prevent breakage. By referring to the diagram of this apparatus (Figure 3), it may be noted that a glass bead is placed in the neck of the centrifuge bottle. The slight imperfection at point of contact between the bead and the neck of the centrifuge bottle acts as a filtration mechanism.

A study of previous swelling tests conducted by Jayme (26, 49) indicates that an important part of the procedure in conducting this type of swelling test is maintaining a rigid control of the rate and time of centrifuging. It is further noted that an acceleration of 800 times the acceleration of gravity for a period of ten minutes permits good reproducibility. This information is thus adopted in determining the centrifuging conditions for the above swelling tests.

The revolutions per minute, η, at which the centrifuge should be operated to give 800 g. may be determined by substituting the appropriate values in the following formula

$$a \text{ (acceleration)} = \left(\frac{2\pi\eta}{60}\right)^2 \cdot r$$

where $\frac{2\pi\eta}{60}$ = angular velocity

r = radius of centrifugal arm which was 15 cm. in this study.

Substituting these values gives

$$800 \text{ g.} = \left(\frac{2\pi\eta}{60}\right)^2 \cdot 15$$

$$\eta^2 = 4{,}770{,}700$$

$$\eta = 2{,}184$$

Thus, in the work in question, 2184 r.p.m. was equivalent to 800 g. acceleration. For purpose of ease of operation, 2200 r.p.m. setting was used in preference to 2184 inasmuch as the tachometer was calibrated in increments of 100 r.p.m. The centrifuge period was 10 minutes. The weight of the pulp slurry sample was permitted to vary from 0.16 to 0.26 g. oven dry.

e. Filtration Resistance. At each beating or repulping cycle, samples of the stock at the end of the beating period were taken from each of two beaters for determining filtration resistance. In addition in the initial or Cycle 0 samples were taken also after the five-minute slushing period. The stock sampled for filtration resistance was at a consistency of approximately 1.57%. It was stored overnight at 35-40°F. and then dispersed in the standard British disintegrator for 15 minutes and then evaluated for filtration resistance.

The filtration resistance was measured employing the method described by Ingmanson and Whitney (44). The stock was evaluated by this method for the purpose of determining the effect of repulping on the filtration resistance of the pulp and the components of resistance--specific surface, specific volume, and pulp compressibility.

Filtration resistance is of value in predicting drainage rates and therefore should be useful as a criterion of how well a stock will drain on the machine. The filtration resistance--ease of water removal--is somewhat analogous but far more precise than the freeness-strength property relationship. Studies carried out have shown that there is a direct relationship between filtration resistance and water removal under a given set of conditions. For the purposes of this study, filtration resistance is reported as the resistance to water drainage expressed in centimeters per gram for a given frictional pressure drop. The filtration resistance per se has no relation to strength properties; however, the components of resistance are related to sheet properties. The specific surface may be defined as the total external surface area. This is the area which the water in the pulp slurry contacts as it flows by in draining on the wire or cylinder. The greater the specific surface, the greater the area for potential bonding. Normally, it is expected that the greater the specific surface, the greater the sheet strength. Also the greater the specific surface, the greater the filtration resistance; a first approximation is that the filtration resistance increases as the square of the specific surface. The specific volume is the hydrodynamic specific volume. It is the volume of fiber and immobilized bound water which is denied to or resists the flow of water; thus it is related to the degree of swelling. Therefore, an increase in swelling is normally associated with an increase in specific volume. Compressibility is the apparent bed density of the wet pad at a given compacting pressure.

The filtration measurements were based on the use of a well-established constant rate technique.

The filtration apparatus consisted of a 3-1/8-inch diameter lucite tube surmounted by a six-inch diameter constant head section. The septum was a 150-mesh screen backed by a 65-mesh screen and supported by a perforated brass disk. The pressure drop across the septum was negligible in comparison to the pressure drop over which the filtration resistance was evaluated.

The slurry was introduced into the filtration tube at approximately 0.01% consistency. The flow rate was maintained at a constant value and the pressure drop across the bed was measured directly by a strip recorder. Then, the filtration resistance was calculated by the relationship,

$$\underline{R} = \frac{A^2}{\mu \underline{q}^2 \underline{c}} , \quad \frac{\Delta P_f}{\theta} ,$$

where $\underline{R}$ = average specific filtration resistance, cm./g.

$\underline{A}$ = area of filtration, 49.5 sq. cm.

$\underline{P_f}$ = frictional pressure drop across the filter bed, dynes/sq. cm.

μ = filtration viscosity, poises

$\underline{q}$ = volumetric flow rate, cc./sec.

$\underline{c}$ = mass of fibers in filter bed per unit volume of filtrate, g./cc.

θ = total time of filtration, sec.

Compressibility measurements were made on the pad formed during the filtration runs by mechanically loading the pad with successive weights and measuring the pad thicknesses with a cathetometer. The values for compressibility and filtration resistance were used in a rectified Kozeny-Carman relationship to calculate specific surface and specific volume. Thus, filtration and compressibility are determined by essentially direct measurement, whereas specific surface and specific volume are calculated identities.

4. Preparation and Evaluation of Handsheets

a. Preparation of Handsheets. As previously mentioned, at each cycle of repulping all the stock (except that used for stock tests) at the end of each beter run, was made up into handsheets with a ream weight of approximately 45 pounds (25x40--500). Two methods (a) open white water system and (b) closed white water system were employed; however, the bulk of the handsheets were made using the closed white water system as only those sheets were used in subsequent repulpings.

(a) Open white water system

The handsheets made with an open white water system were made in accordance with Institute Method 411-3 using British sheet mold. It may be noted that this method includes air drying of the sheets in drying rings for 24 hours in an atmosphere maintained at 50% relative humidity and 73°F. A total of thirty handsheets were made by this method per cycle. A proportionate number of sheets were made from each beater run at each cycle but since the number of beater runs decreased with each repulping cycle, the proportionate number of sheets made from each beater run increased with each repulping cycle.

(b) Closed white water system

The stock remaining at the end of each beater run, after removal of the samples for British handsheets, was drained into a

15-gallon earthenware crock, diluted to 0.32% consistency and stirred with a Lightnin' mixer. Volumes equivalent to 1.88 g. were withdrawn and made into 8-inch circular handsheets on a Schopper (Rapid Köthen) sheet mold equipped with a circulating white water system. The sheet mold and auxiliary equipment is shown in Figure 4. The procedure used was as follows: The white water reservoir was filled with fresh water, the circulating pump started and with valves 2 and 3 closed, valve 1 is set so that the white water is pumped into the lucite deckle box through a series of small orifices placed around the inside periphery of the deckle box just above the 60-mesh wire. When the liquid level was approximately 1 inch above the wire, approximately 590 ml. of the 0.32% consistency stock was added. The white water was allowed to run into the deckle box until a level equivalent to 6-1/2 liters was reached. This gave a deckle box consistency of approximately 0.03%. After the 6-1/2 liters were in the deckle box, valve 1, which is a four-way valve, was set to cut off the water and permit the air to be pumped into the deckle box through the small orifices so as to obtain adequate mixing of the stock. Aeration was permitted for five seconds and then cut off by opening valve 2 and adjusting valve 1 so that air is pumped out of the leg or chamber under the deckle box. After all the water had been drained out of the deckle box, air was drawn through the sheet on the wire for 5 seconds, after which valve 1, which shuts off the air and circulates the white water in the circulating system, was turned to the neutral or off position. The thus-formed sheet was

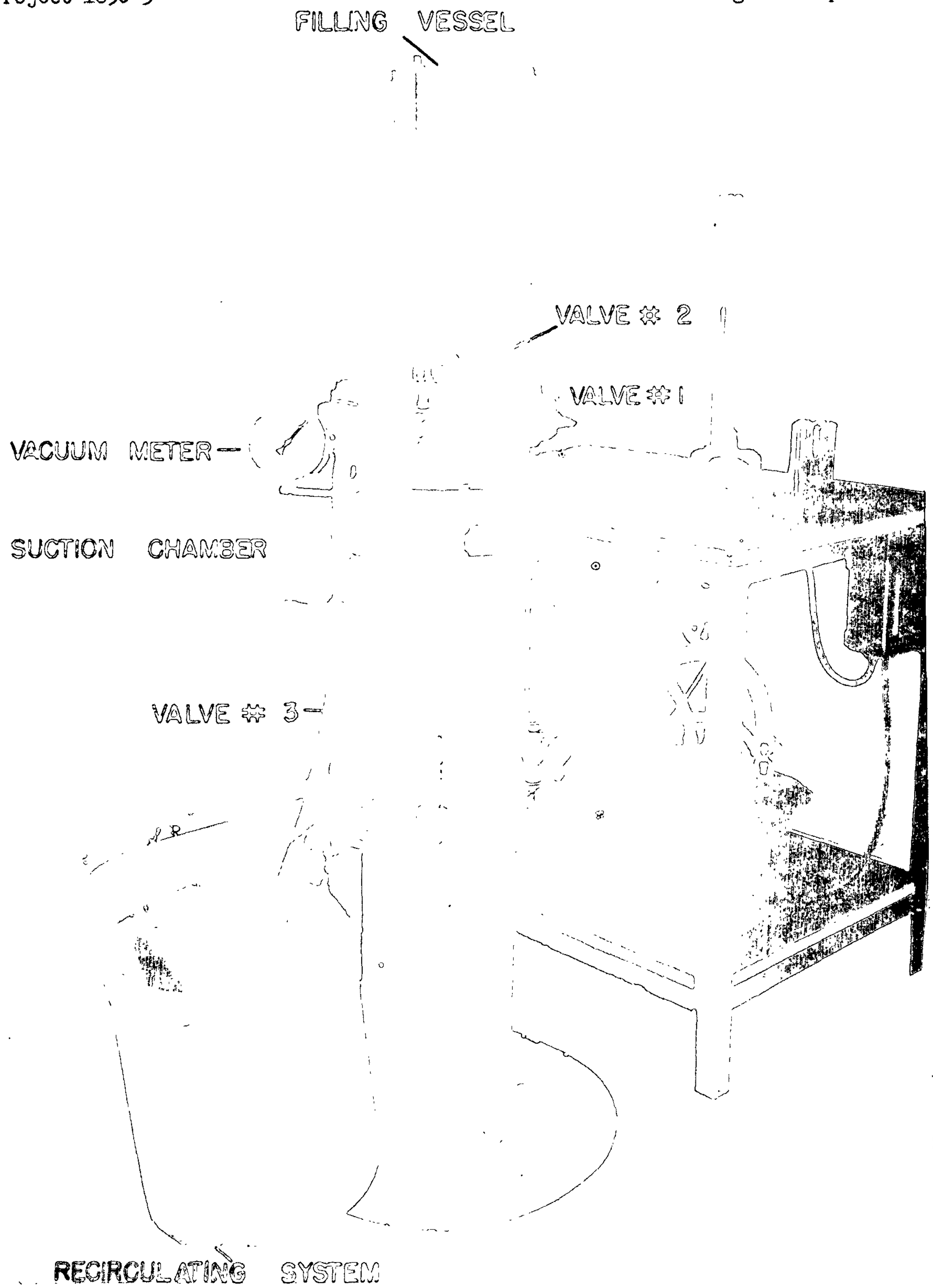

Fig. 4. Schopper (Rapid Köthen) Sheet Machine

covered with a new blotter and couched from the wire. As each sheet was couched from the wire it was covered with a new blotter and when four sheets had been so couched and covered they were placed in a hydraulic press with an extra blotter top and bottom. All four sheets were pressed together using 50 p.s.i. for five minutes, and then dried on a steam-heated (250-260°F.) drier. The handsheets were dried between blotters and the drier canvas. The blotter was between the handsheets and the drier. Each sheet was so dried for 5 minutes.

Inasmuch as it was necessary to fill the white water reservoir with fresh water, the first few sheets were made with white water which had not reached equilibrium in so far as the fines were concerned. In order to determine when the fines in the white water reached a constancy, periodic checks were made of the consistency of the white water at the start of the sheet-making operation. The results obtained for Cycle 0 are given below:

No. Sheets Made	Consistency of White Water, %
1	0.0006
10	0.0011
15	0.0018
20	0.0016
25	0.0017

On the basis of the above results it appears that fine constancy has been attained after about 15 sheets, and at a fine consistency in the neighborhood of 0.0016-0.0018%. On the basis of these results the first twenty-five handsheets were not used for test purposes although it was necessary in certain instances to include them in the furnish used in the next repulping cycle. It may be seen from the data collected later in this study (See Table II) that the fiber content in the white water apparently did not reach a constancy until after more than 25 handsheets had been made, the consistency at the end of the handsheet making being in the order of 0.0030%.

It should be mentioned that the white water reservoir did not have the capacity to permit the accumulation of all the white water and it was necessary to use a small sump pump to remove the excess. It functioned in the following manner. At the start, the white water reservoir was filled with fresh tap water. The water for the mold was drawn from this tank in preparing the first sheet and mixed with approximately 590 ml. of stock; thus, when the sheet was formed, the water draining back into the reservoir exceeded the capacity of the reservoir by an amount equivalent to the water added with the stock, in the neighborhood of 560-580 ml. The sump pump was located so that it handled only the excess and thereby maintained a constant level in the reservoir after the formation of each sheet. The overflow naturally carried with it some of the fines and it has been calculated that approximately 1% stock in the form of fines was lost in this manner.

b. Testing. Prior to testing, all the handsheets were preconditioned for 24 hours at 35% relative humidity and then conditioned for at least 48 hours in an atmosphere maintained at 50 $\pm$ 2% relative humidity at a temperature of 73 $\pm$ 3.5°F. After the prescribed conditioning, the handsheets at each repulping cycle were evaluated for basis weight, caliper, density, bursting strength, tensile strength, stretch, Elmendorf tearing strength, stiffness, fold endurance, porosity, zero span tensile, bonding strength and bonded area. The handsheets made with the open white water system were tested for the same properties and in the same manner as the handsheets made with the closed white water system; however, the results are reported separately. For a given white water system, a proportionate number of the test sheets making up the test samples came from each of the beater runs made at that particular repulping cycle so that each beater run had an equal chance in so far as representation in the test data is concerned. The procedures used for the various tests were as follows:

(1) Basis weight.

Basis weight expressed as the weight in pounds per ream (25x40--500), was determined by trimming 20 handsheets to 5-1/2 x 5-1/2 inches and weighing on an analytical balance to the nearest 0.001 g. and computing ream weight from the weight of the handsheets.

(2) Caliper

One caliper measurement was made on each of the 20 sheets used for basis weight determinations using a Cady micrometer.

(3) Apparent density

The apparent density was calculated from the caliper and basis weight data by dividing the basis weight by the caliper in points.

(4) Bursting strength

Bursting strength tests were performed with a motor-driven "Model C" Mullen tester equipped with a 100-pound gage and also with a special attachment for controlling the clamping pressure on the specimen.

Two test readings were obtained on each of five specimens. On each specimen, one test was obtained with the diaphragm pressure applied to the felt side and one test with the pressure applied to the wire side. The clamping pressure was set at approximately 650 pounds. The ten readings were averaged to give the reported value.

(5) Tensile strength

One test reading was obtained using a Baldwin-Southwark Universal tester on each of ten 3 by 1-inch specimens. The rate of loading was 60 pounds per minute. The average of the ten readings is reported to the nearest 0.1 pound per inch width.

(6) Stretch

Stretch measurements were made on each of ten specimens. The stretch determinations were determined at the same time the tensile measurements were made. The calculated values for the ten readings were averaged and recorded as percentage stretch.

(7) Tearing strength

Tearing strength as measured by the Elmendorf tear tester is the average force in grams necessary to tear a single sheet of paper after the tear has been started. It is, therefore, a measure of the internal tearing resistance rather than the initial or edge-tearing resistance. Four strips of paper 3 by 2.5 inches are held taut between parallel jaws of the instrument. A tear 0.81 inches in length is made at the bottom of the sheets at a right angle to the top of the jaws. The pendulum is then released and the paper torn a distance of 1.69 in. The force in grams necessary to tear the four sheets is recorded and recalculated as the grams of force necessary to tear a single sheet.

The Elmendorf tear tester was used to obtain tear results using 5 specimens of four thicknesses each. The specimens were inserted in the machine such that the wire side faced the machine for the open system sheets and the felt side faced the machine for the closed system sheets. Readings were recorded to the nearest 1.0 gram per sheet.

(8) Fold endurance

Fold endurance tests were made with a Schopper fold tester as directed in Institute Method 513. Ten handsheets were tested at each cycle of repulping. The individual readings were averaged to give the recorded fold endurance value.

(9) Porosity

Porosity measurements were made on a Gurley densometer. Institute Method 514 was used. Two determinations were made on each of five handsheets per cycle of repulping. Half the specimens were tested with the wire side up and the other half with the wire side down. The recorded values are the average of the individual readings in terms of seconds per fifty cubic centimeters of air.

(10) Zero span tensile

Zero span tensile which is supposedly a measure of the tensile strength of the individual fibers was performed using a special zero span tensile holder in the Baldwin-Southwark Universal tester. Ten handsheets per repulping cycle were tested. The specimen width was 0.591 inch and the rate of loading was 60 pounds per minute. The recorded values are the averages of the individual readings in pounds per 1.0 in. width. The individual readings were recorded to the nearest 0.1 pounds.

(11) Bonding strength

The transverse bonding strength of the handsheets at various levels of repulping was determined with the Institute's VVP (velocity-viscosity product) tester (54, 55). In this test the transverse bonding strength is measured in terms of the force necessary to rupture or delaminate the board. The magnitude of the force required to cause rupture is dependent on both the strength of the fiber-to-fiber bond and the number of such bonds. It is therefore not an independent measure of either of these quantities. The instrument consists essentially of a pair of steel wheels having finely ground surfaces on which are applied films of a constant known thickness of a Newtonian oil of known viscosity. The wheels are caused to roll over the test specimen under a known load with constant acceleration and the instantaneous speed is electrically recorded. Two end-points are recorded. One of these is the point at which the paper blisters due to the onset of internal failure. As the paper is subjected to higher and higher force normal to its surface the blister becomes more severe until complete rupture occurs. This is the other end point which is noted. It has been found experimentally that the product of the velocity of the wheels at the blister point and at complete failure and the viscosity of the oil (assuming it is Newtonian) is a constant for a series of samples of the same paper. The results are recorded in kilopoise cm./sec. at the points of blister and rupture. The average values recorded represent the average of 10 tests.

(12) Bonded area

The strength properties of a sheet of paper are dependent to a large extent on the strength with which the fibers, which make up the sheet, are bonded together. The procedure used in this study for determining the bonded area of the handsheets after various repulping cycles involved the following steps:

a. Preparation of an unbonded sheet from the same fiber furnish as the one on which the bonded area is desired.

b. Measure the transmission and reflectance properties of the bonded (regular handsheet) and the unbonded sheet.

c. Utilize Kubelka and Munk theory and its application to light reflectance and transmission of bonded and unbonded fibers in computing the bonded area of the test handsheets.

The bonded sheets used in this determination were selected from the test handsheets made at each level of repulping using the closed white water system. Unbonded sheets were prepared at each level of repulping using the same furnish as was used in the bonded test handsheets. The procedure used was as follows: Up to the point of couching, the sheets were formed on the Schopper sheet mold in the same manner as the test sheets; however, the treatment from there on differed markedly. The reason for this is that if the water in a sheet, before any drying takes place, is replaced by a nonpolar liquid such as benzene, the fibers will not be bonded together on drying. In order to replace the water in the water-formed handsheet on the wire, the suction under the wire was shut off, the deckle box carefully raised and a clean blotter was placed over the formed wet sheet. The deckle box was closed and 500 ml. of acetone slowly poured into the deckle box, the suction applied so as to drain the acetone through the blotter and the wet sheet. Two additional "acetone washings" were carried out each using 500 ml. of acetone. Following the acetone "wash," three additional washes were carried out in the same manner each employing 500-ml. portions of sodium-dried benzene. The thus "washed" sheets were carefully removed with their blotters and then transferred to new blotters. The sheets (on the new blotters) were then dried in a calcium chloride and paraffin desiccator for 24 hours. Note that the above procedure is generally referred to as the butanol method when butanol is used in place of the sodium-dried benzene.

Investigations carried out in the area of the bonding of paper fibers (56) have shown that the scattering coefficient of a sheet of paper may be used to define the bonded area according to the following equation:

$$\frac{s' - s}{s'} \times 100 = \text{per cent bonded area} \qquad (1)$$

where s = scattering coefficient of bonded sheet

s' = scattering coefficient of unbonded sheet

In applying the above relationship to paper, two assumptions are made. These are:

1. The handsheets prepared by the benzene method are complately unbonded.

2. The scattering power or scattering coefficient is linearly proportional to the unbonded area.

With the acceptance of these two assumptions, attention may be directed to the measurement of the scattering coefficient of the bonded and unbonded sheets. From the theory of reflectance and transmission of light by paper, it can be shown that the scattering power, sW, for a given sheet may be computed from a knowledge of the reflectance, R_∞, of a pad of infinite thickness and the total transmission of the sheet, T. These two latter identities were determined conveniently using the G. E. recording spectrophotometer. Reference is then made to the appropriate computational chart and the value of sW corresponding to the above determined values of R_∞ and T read directly. The scattering coefficient may be determined by dividing the scattering power sW by the basis weight W.

In the present study five handsheets per cycle of repulping were evaluated for reflectance and total transmission. The basis weight was accurately determined on conditioned samples which were used for reflectance and transmission determination. A circular knife edge punch was used to cut the samples and the diameter was measured with a microscope. Basis weight was calculated in g./sq. cm. for an area of 8.1221 sq. cm.

D. DISCUSSION OF RESULTS

As previously mentioned, the purpose of this study was to determine the effect of progressive repulpings on the physical properties of the fibers and the sheets made therewith. It is common knowledge throughout the industry that once a fiber has been pulped, refined, made into a sheet and dried, that upon reuse it has not been possible to develop the fibers to the same strength level as they possessed as virgin fibers. Although this has been general knowledge for many years, there has been very little attention focused on the problem and there are little data available as to the properties which are affected and to what extent. Furthermore, there is no information available as to whether the adverse influence on strength properties is a function of the number of times repulped or whether the entire change takes place between the initial pulping and the first repulping cycle. Consequently, the main objective of this phase is to determine what properties of the fiber and sheet are influenced by repulping and to what extent.

The repulping phase carried out was designed to simulate as nearly as possible the papermaking environments which a fiber would experience from the virgin state through six repulpings with the hope that these simulated conditions would influence the fiber and the sheet made therefrom in the same manner as in actual commercial operation. Once we know which properties are influenced by the refining and papermaking operations, attention may be focused on ways and means of determining the mechanisms through which the strength adversities are manifested and how they can best be minimized or eliminated.

It should be recalled that the study was designed using normal yield, southern, softwood unbleached kraft as the starting raw material. The procedure used consisted in refining the virgin kraft pulp in a laboratory Valley beater to a given freeness level (615 ± 15 cc. Schopper-Riegler) and then making up all the stock into standard weight handsheets. With the exception of a few test sheets all the stock was made up into standard weight handsheets on a Schopper sheet machine (Rapid Köthen) equipped with a recirculating white water system and dried on a steam-heated drier drum. Tests were made of the properties of the stock and also of the handsheets. The handsheets so made, which were not used for test purposes, were used as the fibrous raw material for the first repulping. The first repulping and all subsequent repulpings were carried out in the same manner as the initial beater-run and sheetmaking operations. The drum-dried handsheets from the first repulping not used for test purposes became the fibrous raw material for the second repulping. This procedure was followed until the fibers had been subjected to six cycles of repulping. Tests were made on the stock before and after refining at each cycle of repulping and the handsheets at each repulping were evaluated for various strength properties.

For purposes of presentation, the results have been divided into 1.- The effect of repulping on fiber properties, and 2.-The effect of repulping on sheet properties.

1. The Effect of Repulping on Sheet Properties

As was mentioned earlier, the stocks in the various repulping cycles were all refined to the same level in terms of Schopper-Riegler freeness. Test handsheets were formed on a mold with an open white water system and subsequently air dried, and also on a mold equipped with a recirculating white water system. The latter sheets were all dried on a steam-heated drier drum to simulate machine conditions. The purpose of the two systems was to compare the effects of fines and mode of drying on sheet properties at the various levels of repulping.

a. Closed white water system, drum drying.

The results of the evaluation of the handsheets made at the various levels of repulping are given in Table II and graphically illustrated in Figures 5-9. It may be seen from the data in Table II and Figure 5 that the bursting strength decreased rapidly with the number of times the stock was repulped. After six repulpings, the resulting bursting strength was approximately 30% lower than on the initial refining. In addition, the slope of the curve in Figure 5 indicates that there is a rather rapid decrease in bursting strength with number of repulpings for the first two cycles. After the fourth

TABLE II

STRENGTH PROPERTIES OF HANDSHEETS AT VARIOUS LEVELS OF REPULPING

(Closed System, Drum Dried)

Number of times repulped	0	1	Difference	2	Difference	3	Difference	4	Difference	5	Difference	6	Difference
Properties of handsheets													
Basis weight, lb. (25x40x500)	47.6	46.0		45.9		44.3		45.5		44.9		45.7	
Caliper, points	5.2	4.3		4.8		5.2		5.4		5.5		5.6	
Apparent density	9.2	8.7	-5.4	8.5	-7.6	8.5	-7.6	8.4	-8.7	8.2	-10.9	8.2	-10.9
Bursting strength,													
p.s.i.	45.1	41.4		34.7		34.0		33.9		30.7		30.8	
Factor	0.95	0.90	-5.3	0.76	-20.0	0.74	-22.1	0.68	-28.4	0.67	-29.5	0.67	-29.5
Tensile strength,													
Load, lb./in. width	25.7	22.2		19.4		19.0		18.6		18.1		17.9	
Factor	0.54	0.48	-11.1	0.42	-22.2	0.43	-20.3	0.41	-24.1	0.40	-25.9	0.39	-27.8
Stretch, %	3.7	3.3	-10.8	2.8	-24.3	3.1	-16.2	3.1	-16.2	2.9	-21.6	3.1	-16.2
Continued tearing str.													
g./sheet	84	90		95		93		94		97		105	
Factor	1.76	1.96	+11.4	2.07	+17.6	2.10	+19.3	2.06	+17.1	2.16	+22.7	2.30	+30.7

TABLE II--Continued

STRENGTH PROPERTIES OF HANDSHEETS AT VARIOUS LEVELS OF REPULPING
(Closed System, Drum Dried)

Number of times repulped	0	1	Difference	2	Difference	3	Difference	4	Difference	5	Difference	6	Difference
Properties of handsheets--continued													
Taber stiffness													
Unit	2.1	2.0		2.0		1.8		2.0		2.1		1.9	
Factor	.044	0.043	-2.3	0.044	0.0	0.041	-7.0	0.044	0.0	0.047	+7.0	0.042	-2.3
Schopper fold No. of folds	716	573	-20.0	457	-36.2	382	-46.6	329	-54.1	390	-45.5	248	-65.4
Porosity, sec./100 cc.	42	79	+88.1	44	+4.8	48	+14.3	46	+9.5	40	-4.8	35	-16.7
Zero span tensile,													
lb./in.	42.8	41.3		40.1		39.3		36.9		38.9		40.1	
Factor	0.90	0.90	0	0.87	-3.3	0.89	-1.1	0.81	-10.0	0.87	-3.3	0.88	-2.2
Bonding strength, kilopoise cm./sec.													
Blister	339	216	-36.3	212	-37.5	172	-49.3	171	-48.6	136	-59.3	129	-61.9
Rupture	446	314	-24.2	290	-35.0	282	-36.8	235	-47.3	195	-56.3	197	-55.8
Bonded area, %	59.0	58.7	-0.5	57.2	-3.1	54.6	- 7.5	56.2	-4.7	46.3	-21.5	62.2	+5.4
White water solids,* %	.0031	.0030		.0020		.0025		.0028		.0033		.0024	

* Determined at end of sheetmaking operation.

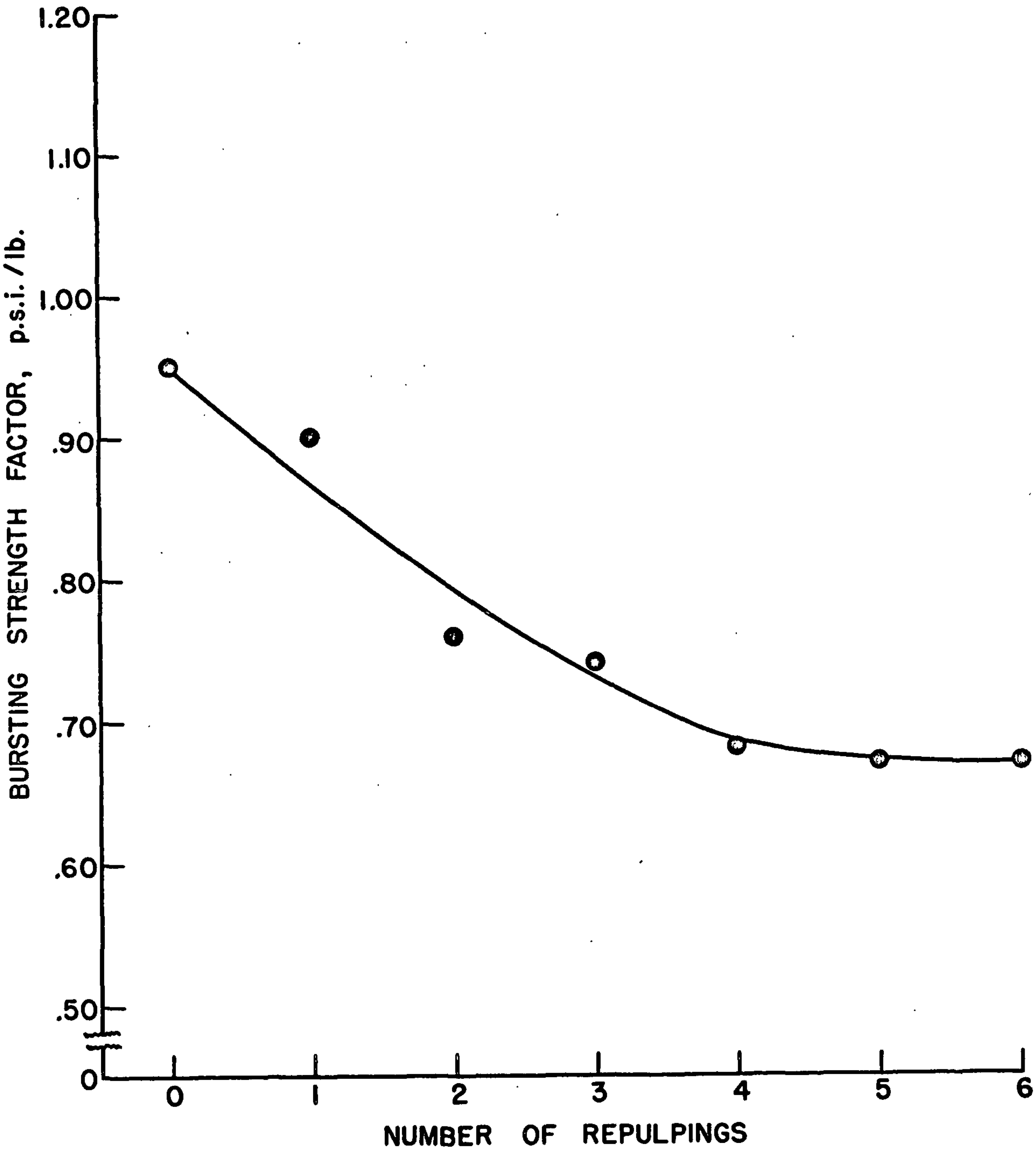

Fig. 5. Effect of Repulping on Bursting Strength

(Closed System, Heat Dried)

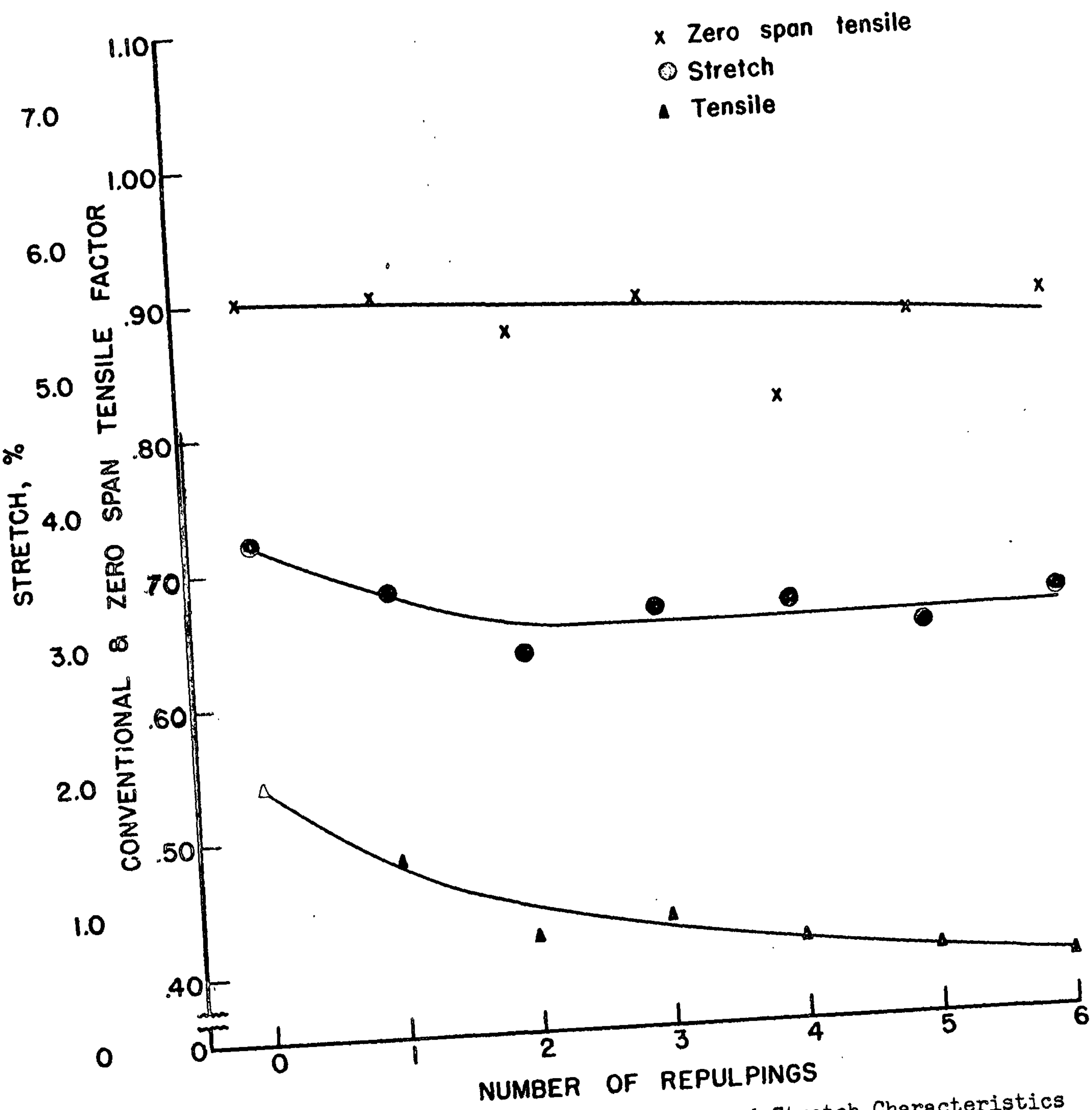

Fig. 6. Effect of Repulping on Tensile and Stretch Characteristics
(Closed System, Heat Dried)

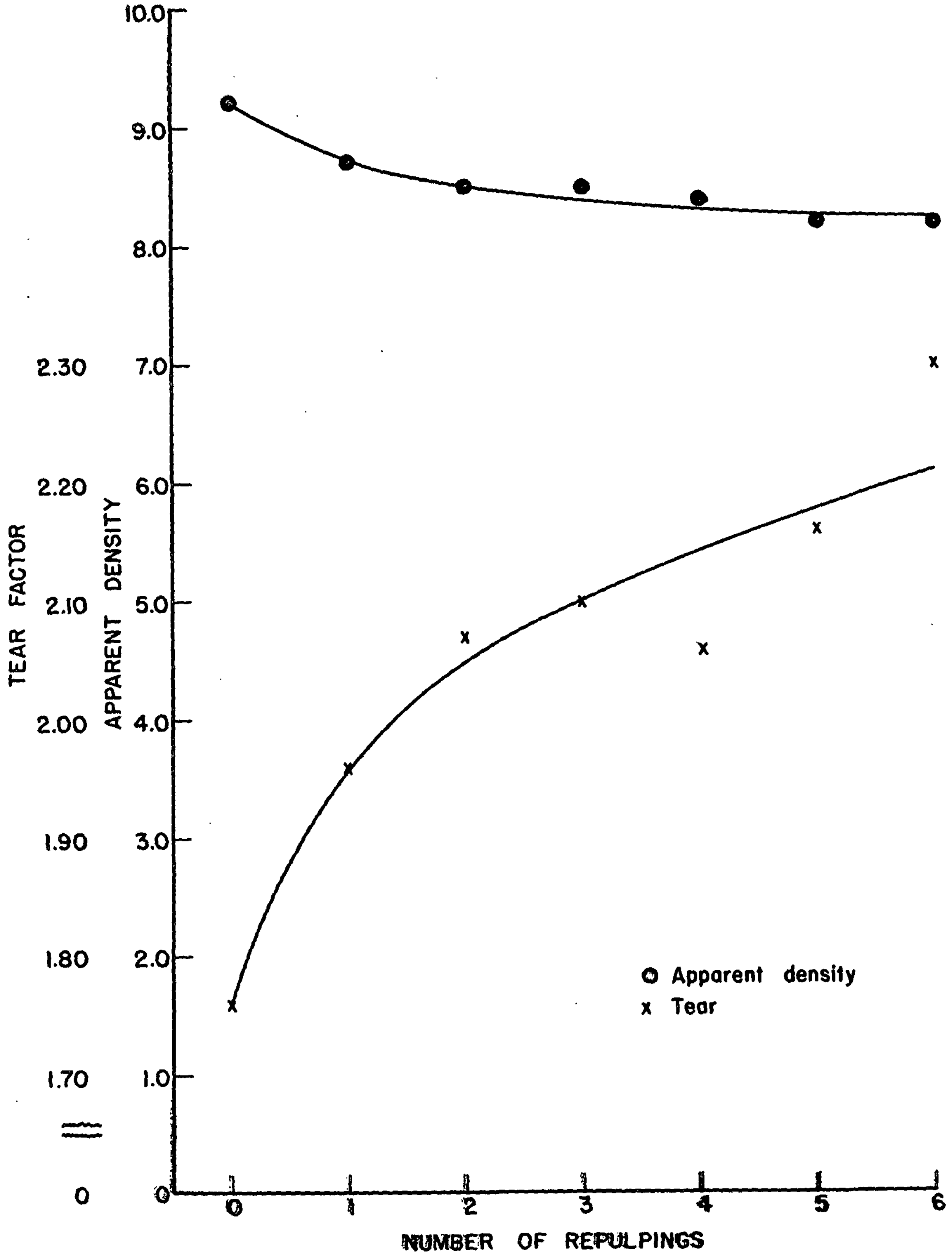

Fig. 7. Effect of Repulping on Apparent Density and Tearing Strength (Closed System, Heat Dried)

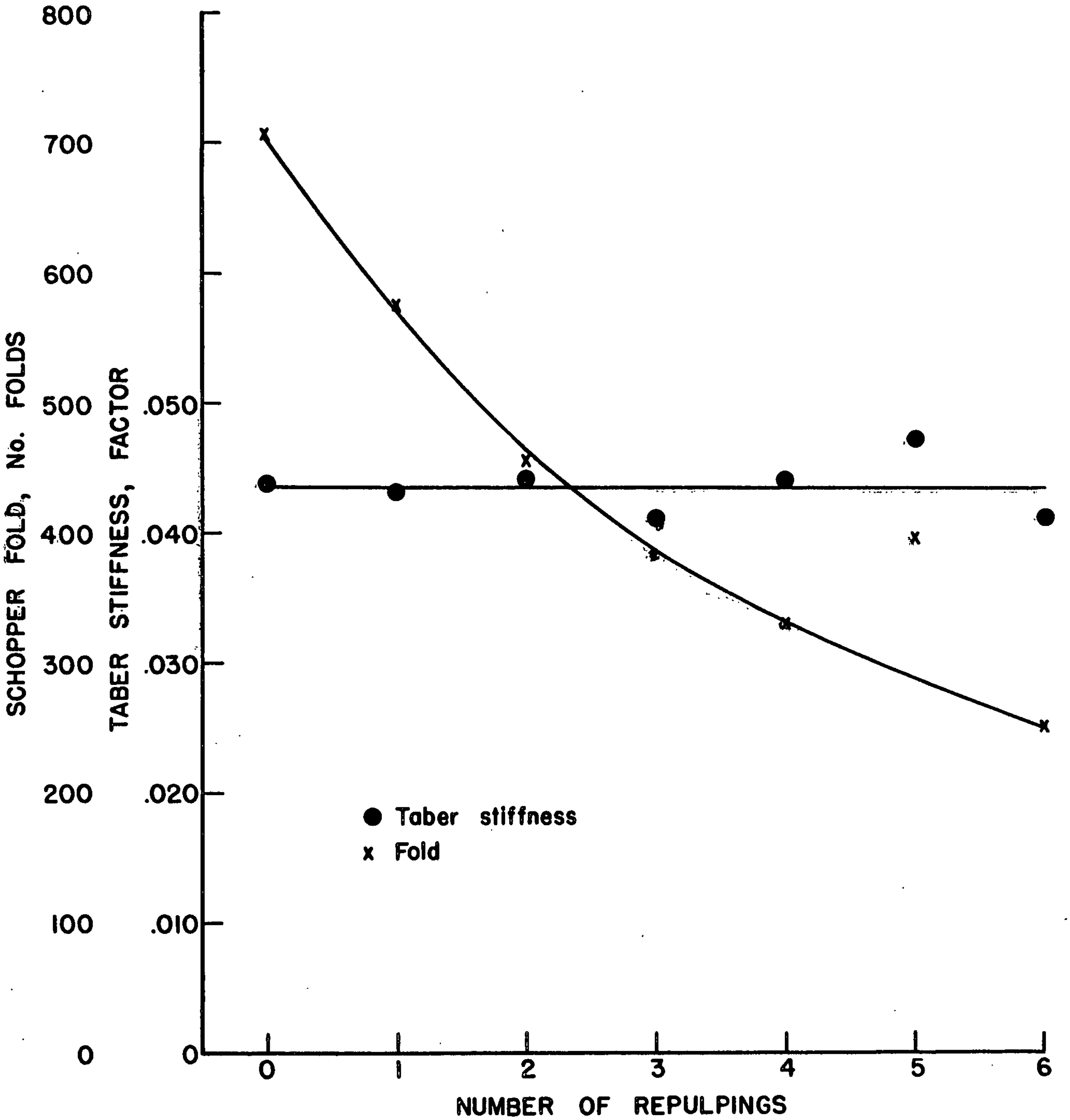

Fig. 8. Effect of Repulping on Fold Endurance and Taber Stiffness
(Closed System, Heat Dried)

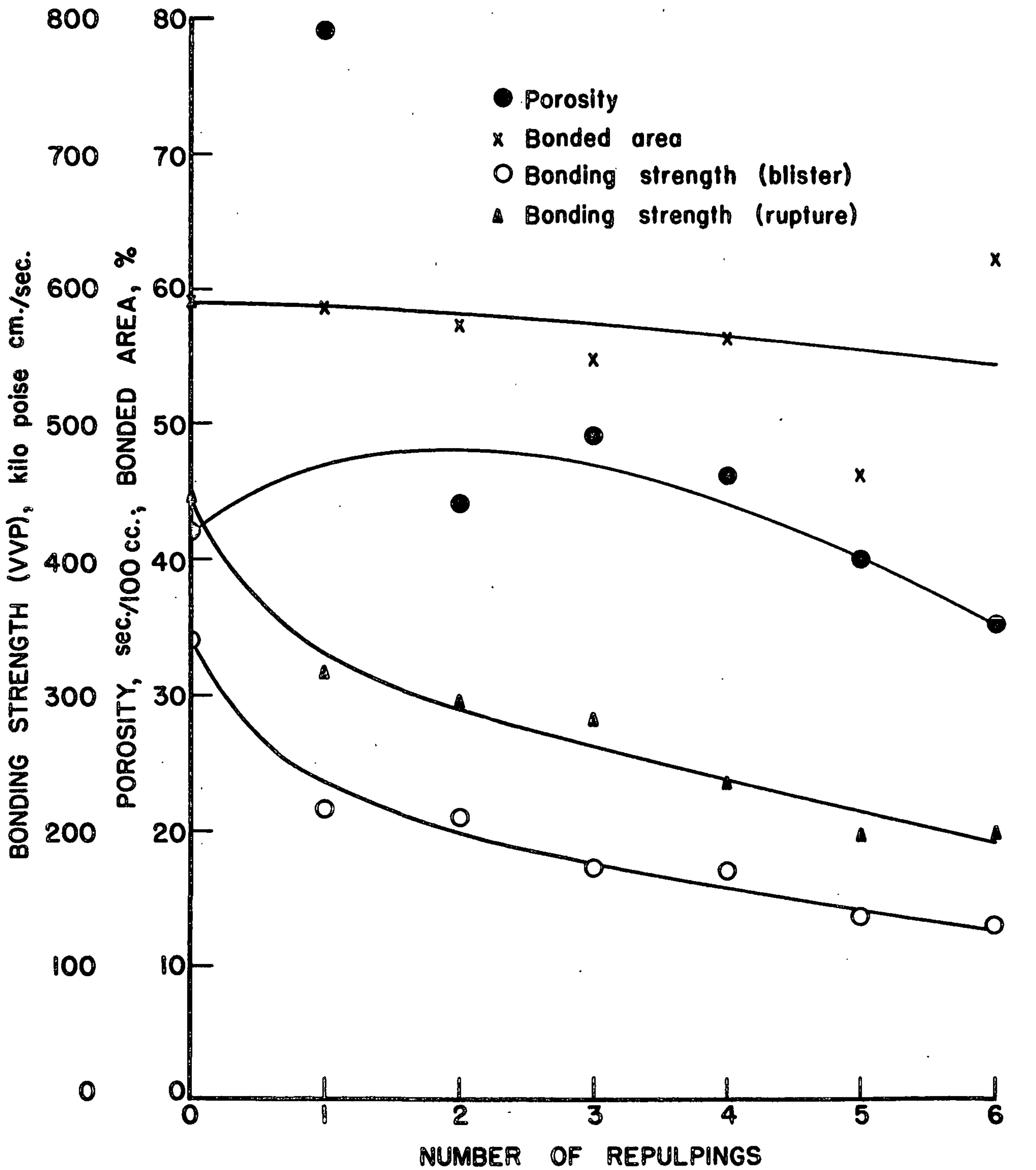

Fig. 9. Effect of Repulping on Porosity, Bonding Strength and Bonded Area (Closed System, Heat Dried)

cycle, there is little change in bursting strength. The bursting strength of handsheets is primarily a function of the tensile and stretch characteristics, as may be seen from the following generalized relationship,

$$P = \frac{2\,T}{R}$$

where P = bursting strength,

T = the tensile strength,

R = the radius of curvature of the diaphragm at the moment of rupture.

The radius of curvature of the diaphragm is a direct function of the stretch characteristics of the sheet, the greater the stretch, the smaller the radius and consequently, the greater the bursting strength.

The effect of repulping on the conventional tensile, zero-span tensile, and stretch characteristics may be seen in Figure 6. It may be noted that the tensile strength of the handsheets decreased progressively with the number of times the stock was repulped. At the end of the sixth cycle the tensile strength was approximately 28% lower than initially. In addition, it may be seen that the rate of change was greatest for the first two repulping cycles. It is generally considered that the tensile strength of a sheet of paper is a function of the strength of the fibers and the strength and number of bonds with which the fibers in a sheet are held together. In the conventional test, the strength observed is a manifestation of both these properties. Thus, the tensile results indicate that one or all of these identities are decreasing. The zero span tensile test was developed for the purpose of measuring the composite strength of the fibers only. There is some question as to the degree to which the test approaches this goal; however, it is generally held that the zero-span tensile measures predominantly fiber strength as distinguished from interfiber bonding strength. The curve for zero-span tensile indicates that there was only a slight decrease in fiber strength with number of times repulped, being of the order of 2--3% except for Cycle 4. These results would indicate that the refining in the repulping cycle was not such that it caused a very marked decrease (such as by fibrillation, mashing or cutting) in the strength of the individual fibers. The stretch characteristics decreased rather sharply for the first and second repulping and then leveled off at about 15-20% loss level.

The results obtained for apparent density--i.e., basis weight divided by caliper--and Elmendorf tearing strength are plotted in Figure 7. It may be observed that the apparent density decreased with increasing number of times the stock is repulped. At the end of the sixth cycle the apparent density is about 10% lower than initially. Since apparent density is a calculated value obtained by dividing the basis weight by the caliper, the higher the caliper for a given basis weight the lower the apparent density. For a given stock under normal conditions the apparent density is an indirect indication of the degree of refinement and hence bonding. The more a given stock is refined, the lower the caliper and the higher the density because of the greater bonding; thus, the apparent density results indicate that there was a decrease in bonding on repulping. The relationship between tear and number of repulpings shown in Figure 7 indicate that under the conditions of the study the tear values increased with number of repulpings. This behavior may at first appear to be at odds with experience in that it is well known that the tearing strength of jute liner made from recovered paper fiber is one of its greatest weaknesses, particularly as compared to kraft linerboard. It should be borne in mind, however, that it is only the machine-direction tear which is weak as the across-machine tear normally is considerably in excess of its competitive grade; thus the question may be posed as to whether the low machine-direction tear is predominantly a fiber characteristic or a machine characteristic. The author strongly suspects that in the case of jute liner the low machine-direction tear is primarily a manifestation of the effect of the cylinder machine on strength properties.

It has been shown (57) that the total energy expended in tearing a sheet of paper is composed of the work required to (a) break individual fibers in tension, and (b) pull individual fibers out of the mesh of fibers. In the latter the energy expended is that required to overcome the drag or resistance due to the bonding of the contiguous fibers. Thus, the total energy required to tear a sheet of paper may be expressed as follows:

$$\text{Total energy} = n_r\, w_r + n_d\, w_d$$

where n_r = number of fibers ruptured in tearing sheet

w_r = average work required to rupture a fiber

n_d = number of fibers pulled from mesh in tearing sheet

w_d = average work required to pull a fiber out of mesh

The total work going into the tearing of a sheet may be represented as 2 Fe where F is the observed tearing force and e is the length of tear. The factor 2 is used because, in order to tear a sheet through a given distance, the tearing force has to be applied through twice that distance. Equating these two energies gives

$$F = (n_r\, w_r + n_d\, w_d)/\ 2e$$

If papermaking stock is beaten to a degree beyond the early stages--i.e., where increased fiber bonding increases tearing strength indirectly by increasing the tightness of the fiber mesh--the addition of an agent which will increase the bonding strength will cause a decrease in the tearing strength. An increase in bonding strength results in an increase in the number of fibers ruptured and a corresponding decrease in the number of fibers pulled out. Since the drag work has been shown (57) to be greater than the rupture work, any decrement in the number of fibers pulled out will cause a decrease in the tearing force. Similarly, the addition of an agent either mechanical or chemical which decreases the bonding strength will result in a decrease in the number of fibers ruptured and an increase in the number of fibers pulled out; thus, the tearing force will increase. As will be pointed out later, the fiber length did not change appreciably, whereas the bonding strength decreased markedly with progressive repulpings. It is believed that this latter is probably responsible for the increase in tearing strength. Further, it may be observed that the tearing strength increased more rapidly during the first and second repulping cycle, which, it will be shown, corresponds to the period of most rapid decrease VVP bonding strength, degree of swelling and apparent density.

The change in Taber stiffness and Schopper fold endurance with number of times repulped may be seen in Figure 8. On the basis of these results it would appear that there is no significant change in Taber stiffness. In any bending stiffness test, there are two factors which have a dominant influence, namely, the inherent material strength of the specimen and its geometry--i.e., moment of inertia. Stiffness may be defined as EI where E is the modulus of elasticity and I the moment of inertia. The more a stock is refined, normally the better the bonding, and the higher is the modulus of elasticity. However, this increase in modulus of elasticity is accompanied by an increase in the density of the sheet and, consequently, a lower caliper which im turn brings about a lower moment of inertia. Thus, it may be reasoned that there are two opposing but not necessarily equal forces working simultaneously. Whether the stiffness increases or decreases with change in refining, etc., depends on which force is the dominant one.

The fold endurance results plotted in Figure 8 show that this property is very sensitive to the changes which the fibers undergo on progressively beating, forming, and drying. The fold endurance strength of the handsheets made from fibers which had been repulped six times was about 65% lower than initially. The fold endurance strength of a sheet of paper is generally considered to be a function of the fiber length, degree of bonding, strain characteristics and moisture content. The fold endurance decreased with repulping, the greatest rate of decrease being in the first two repulping cycles although the change thereafter was appreciable. The present results would indicate that those properties on which fold depends are developed to a progressively lesser extent on repulping.

The results tabulated in Table II for porosity, bonding strength, and bonded area are graphically illustrated in Figure 9. It may be seen that although the results are quite variant, there is a tendency for the handsheets to be slightly more porous--i.e., lower time for air displacement-- with greater number of repulpings. This is in keeping with the observed lower apparent density. The transverse bonding strength as defined by the VVP tester shows that the bonding strengths at the points of blister and rupture both decrease sharply with the first repulping and then at a slower rate with progressive repulpings. The transverse bonding strength is dependent on both the strength of the fiber-to-fiber bonds and the number of such bonds. The decrease in bonding strength after six repulpings amounted to between 55 and 62% for the rupture and blister endpoints, respectively. Because of the dependency of the transverse bonding strength on the number and strength of the fiber-to-fiber bonds it would appear that the effect of repulping is to decrease either or both of these identities.

The effect of the number of repulpings on the bonded area may be seen from the curve in Figure 9. As may be noted, there is a tendency for the bonded area to decrease only slightly with each repulping after the first repulping. These results confirm the trend noted for the VVP transverse bonding strength. On the basis of those two tests, it would appear that not only is the bonded area decreasing slightly with each repulping, but there is strong evidence that the strength of the fiber-to-fiber bond is also decreasing and these two--i.e., slightly lower bonded area and weaker fiber-to-fiber bonds--manifest themselves in the form of lower bursting strength, tensile, stretch, fold endurance and transverse bonding strength but higher tearing strength.

The percentage change in bursting strength, apparent density, tearing strength, tensile strength, Taber stiffness, fold endurance, bonding strength and bonded area are shown in Figures 10 to 13.

b. Open white water system, airdried sheets.

The results of the evaluation of the handsheets made at the

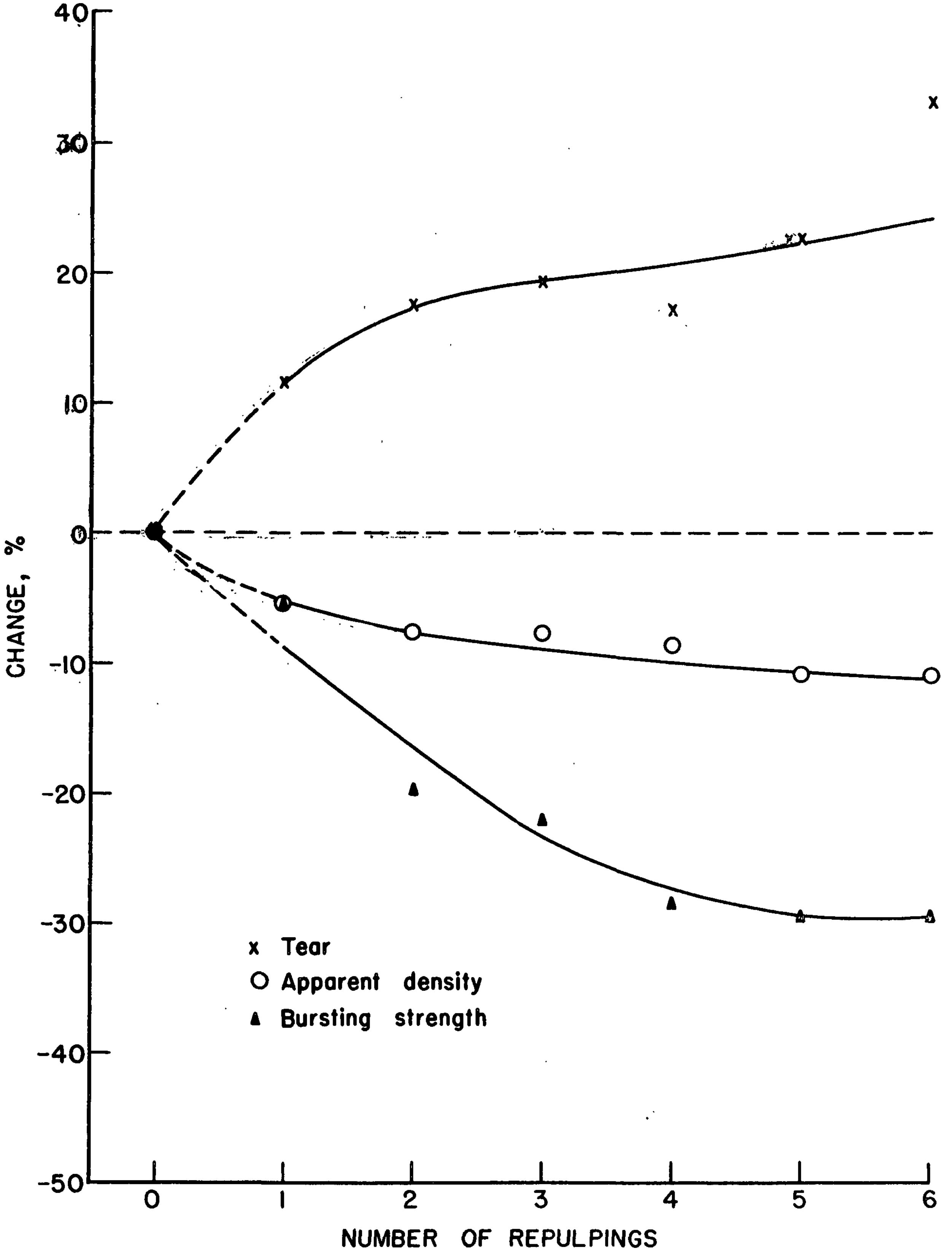

Fig. 10. Change in Bursting Strength, Apparent Density, and Tearing Strength With Repulping
(Closed System, Heat Dried)

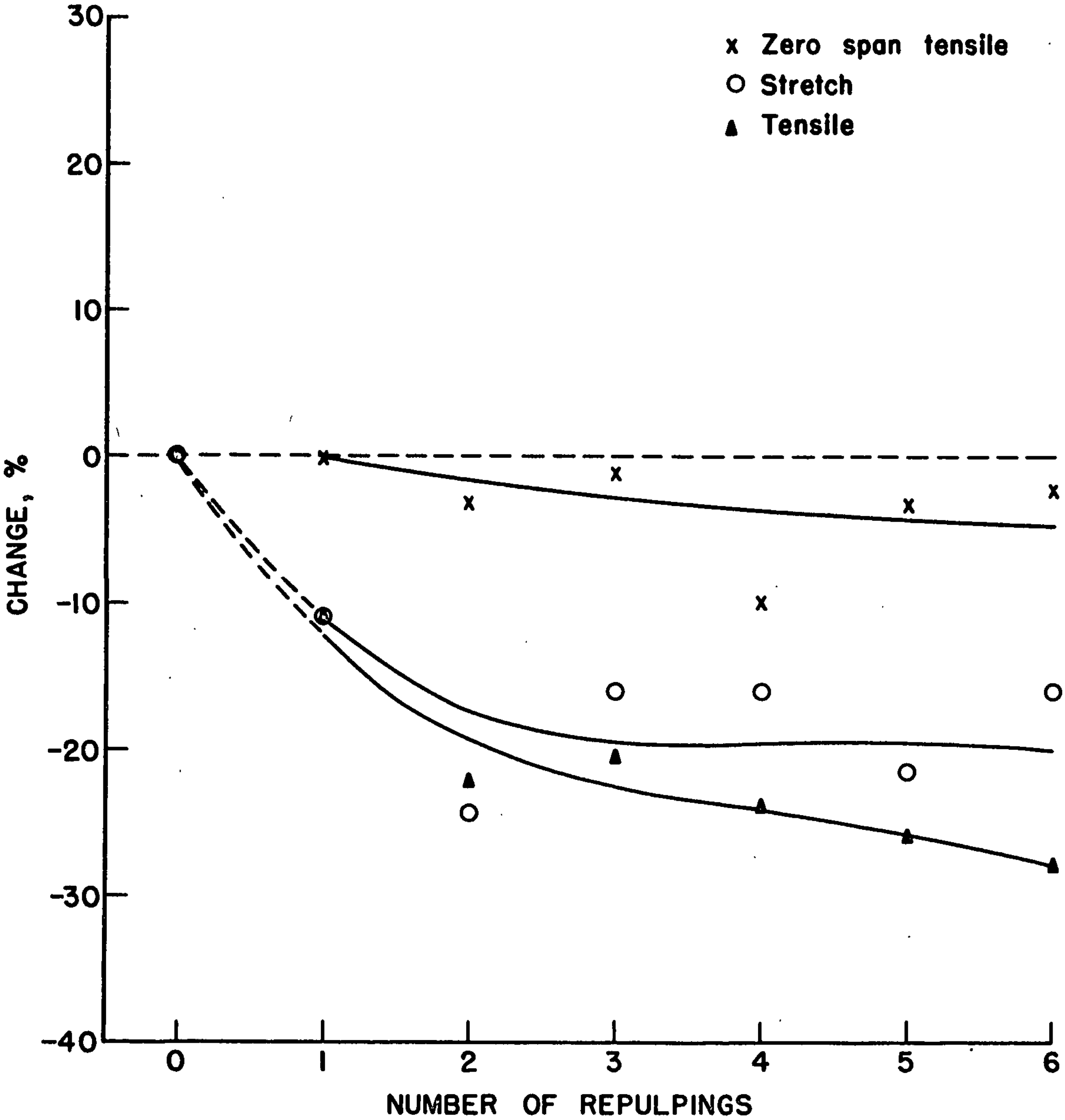

Fig. 11. Change in Tensile and Stretch Characteristics with Repulping

(Closed System, Heat Dried)

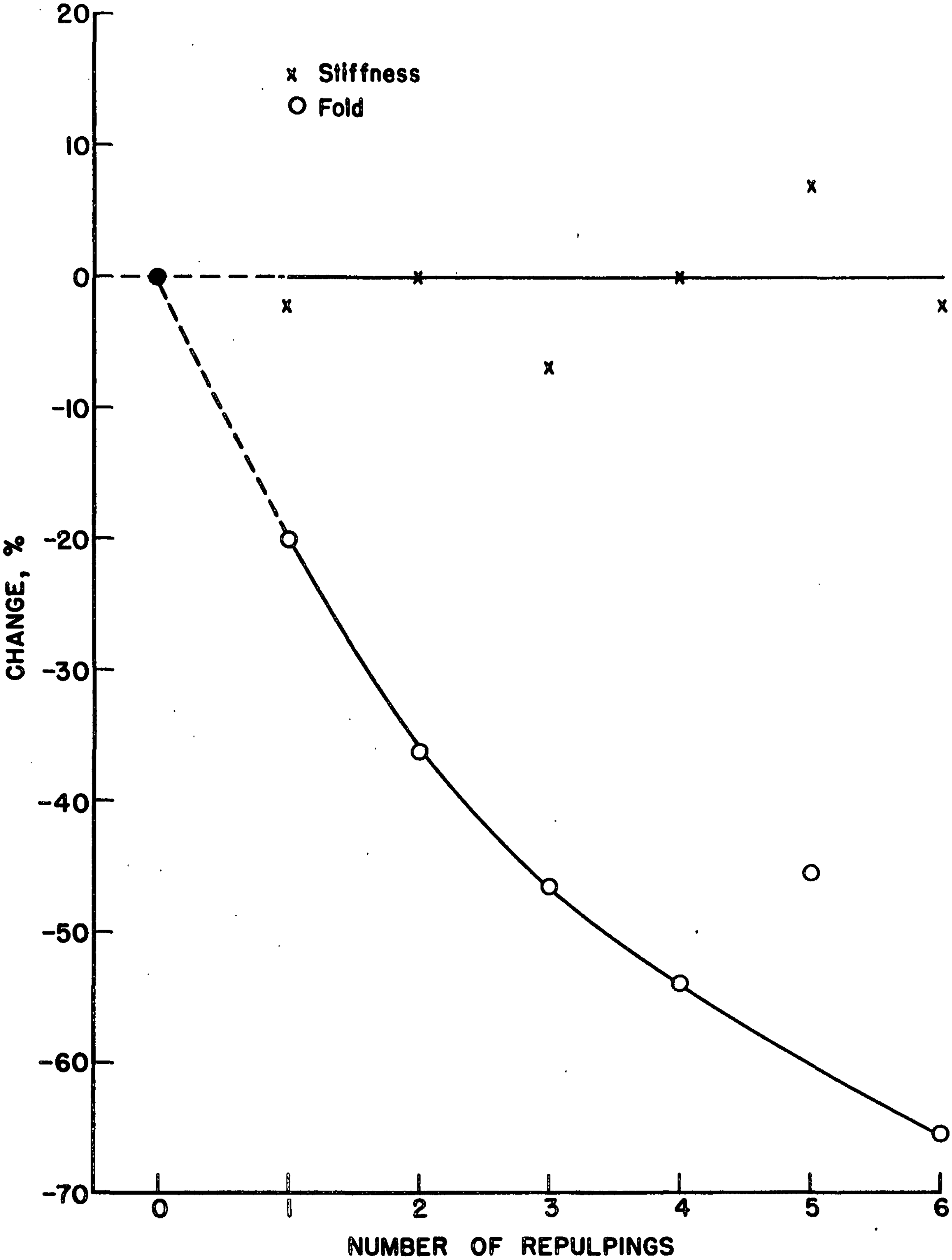

Fig. 12. Change in Fold Endurance and Stiffness Characteristics
With Repulping
(Closed System, Heat Dried)

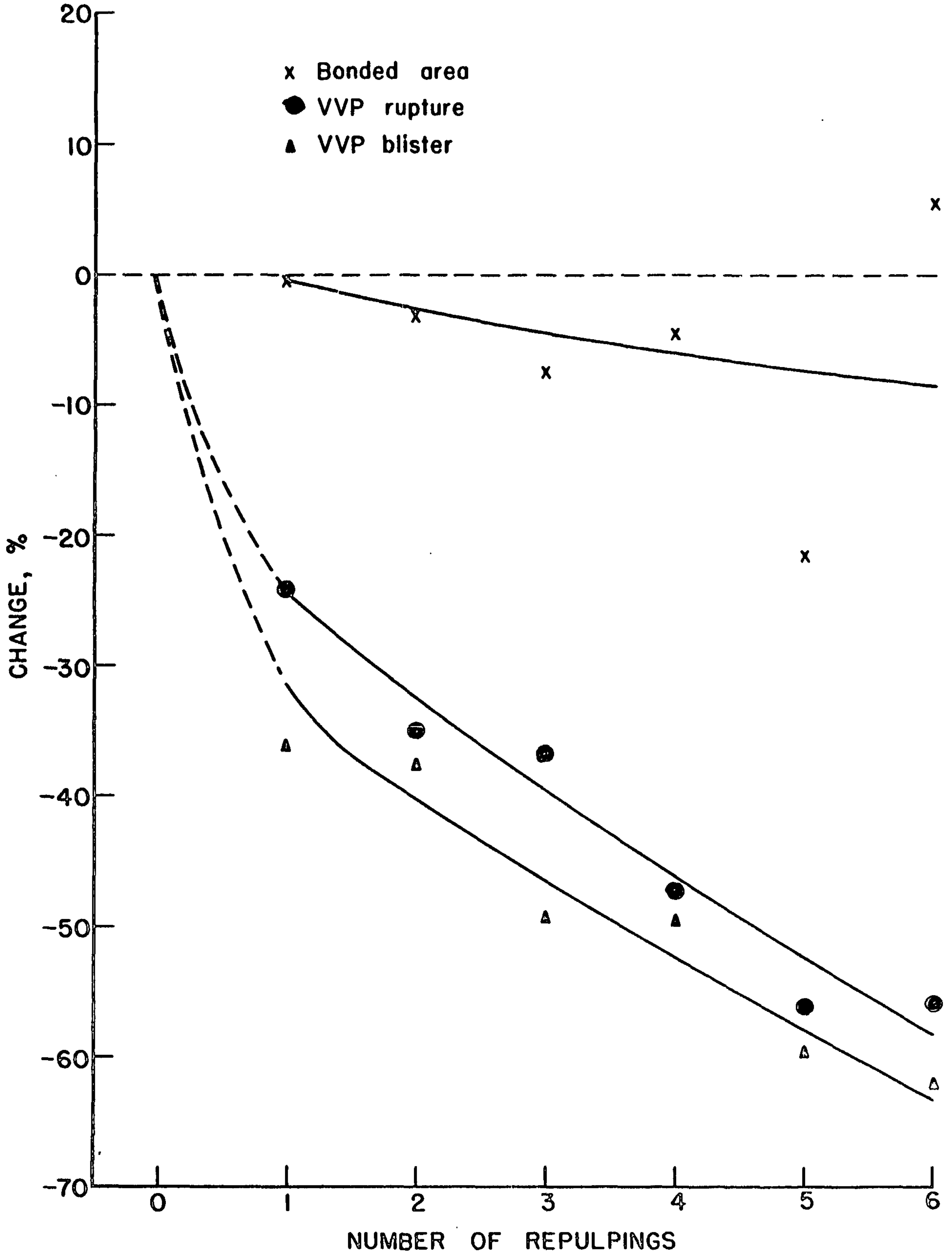

Fig. 13. Change in Bonding Strength and Bonded Area with Repulping
(Closed System, Heat Dried)

various levels of repulping using an open white water system and air drying are given in Table III and graphically illustrated in Figures 14 to 18. The stocks used in preparing the handsheets tested in this section were the same as used in the previous section, utilizing a closed white water system and drum drying. The handsheets tested in this section were prepared in a standard British sheet machine using an open white water system and airdrying the sheets. Thus, the handsheets in this section differ from the handsheets in the previous section among other things in fine content and mode of drying.

It may be seen from the bursting strength data presented in Table III and graphically illustrated in Figure 14 that the bursting strength decreased markedly with increase in number of times the fibers were repulped. The loss in bursting strength after six repulpings was approximately 37%; the most rapid rate of decrease is during the early repulping cycles. These data confirm the behavior observed for the handsheets made with the closed white water system and drum dried. The loss in bursting strength, however, was greater.

The relationships between conventional tensile, zero-span tensile and stretch properties and the number of times repulped may be seen by referring to Figure 15. It may be noted that the conventional tensile strength was decreased progressively with the number of times repulped; after the sixth cycle, it was approximately 30%

TABLE III

STRENGTH PROPERTIES OF HANDSHEETS AT VARIOUS LEVELS OF REPULPING
(Open System--Air Dried)

Number of times repulped	0	1	Difference	2	Difference	3	Difference	4	Difference	5	Difference	6	Difference
Properties of handsheets													
Basis weight, lb. (25x40x500)	45.4	45.1		46.2		46.3		46.2		48.0		47.0	
Caliper, points	4.3	4.5		4.8		4.9		5.0		5.0		5.1	
Apparent density	10.6	10.0	-5.7	9.6	-9.4	9.4	-11.0	9.2	-13.2	9.6	-9.4	9.2	-13.2
Bursting strength													
p.s.i.	47.2	40.4		37.6		34.2		32.3		33.2		30.4	
Factor, lb./lb. fiber	1.04	0.90	-13.5	0.81	-22.1	0.74	-28.8	0.70	-32.7	0.69	-33.7	0.65	-37.5
Tensile strength,													
Load, lb./in	26.2	24.5		20.7		20.7		20.5		20.0		19.2	
Factor	0.58	0.54	-6.7	0.45	-22.4	0.45	-22.4	0.44	-24.1	0.42	-27.6	0.41	-29.3
Stretch, %	3.2	3.0	-6.3	2.9	-9.4	2.6	-18.8	2.4	-25.1	2.5	-21.9	2.6	-18.8
Combined tearing strength													
g./sheet	84	89		96		100		97		106		108	
Factor	1.85	1.97	+6.5	2.08	+12.4	2.16	+16.8	2.10	+13.5	2.20	+18.9	2.30	+24.3
Taber stiffness													
Unit	2.1	2.2		2.2		2.2		2.4		2.5		2.5	
Factor	0.046	0.049	+6.5	0.048	+4.3	0.048	+4.3	0.052	+13.0	0.052	+13.0	0.053	+15.2
Schopper fold, No. folds	599	565	-5.7	472	-21.2	379	-36.7	354	-40.9	467	-22.0	316	-47.2

TABLE III--Continued

STRENGTH PROPERTIES OF HANDSHEETS AT VARIOUS LEVELS OF REPULPING
(Open System--Air Dried)

Number of times repulped	0	1	Difference	2	Difference	3	Difference	4	Difference	5	Difference	6	Difference
Properties of handsheets--continued													
Porosity, sec./100 cc.	30	36	+20.0	33	+10.0	23	-23.3	22	-26.7	21	-30.0	20	-33.0
Zero span tensile load, lb./in	43.0	40.9		42.8		40.4		37.2		35.5		33.8	
Factor	0.95	0.91	-4.2	0.93	-2.1	0.87	-8.4	0.81	-14.7	0.74	-22.1	0.72	-24.2
Bonding strength, kilopoise cm./sec.													
Blister	300	192	-36.0	153	-49.0	170	-43.3	146	-51.0	107	-64.3	92.3	-69.2
Rupture	487	354	-27.3	293	-39.8	268	-45.0	241	-50.5	212	-56.5	175	-64.1

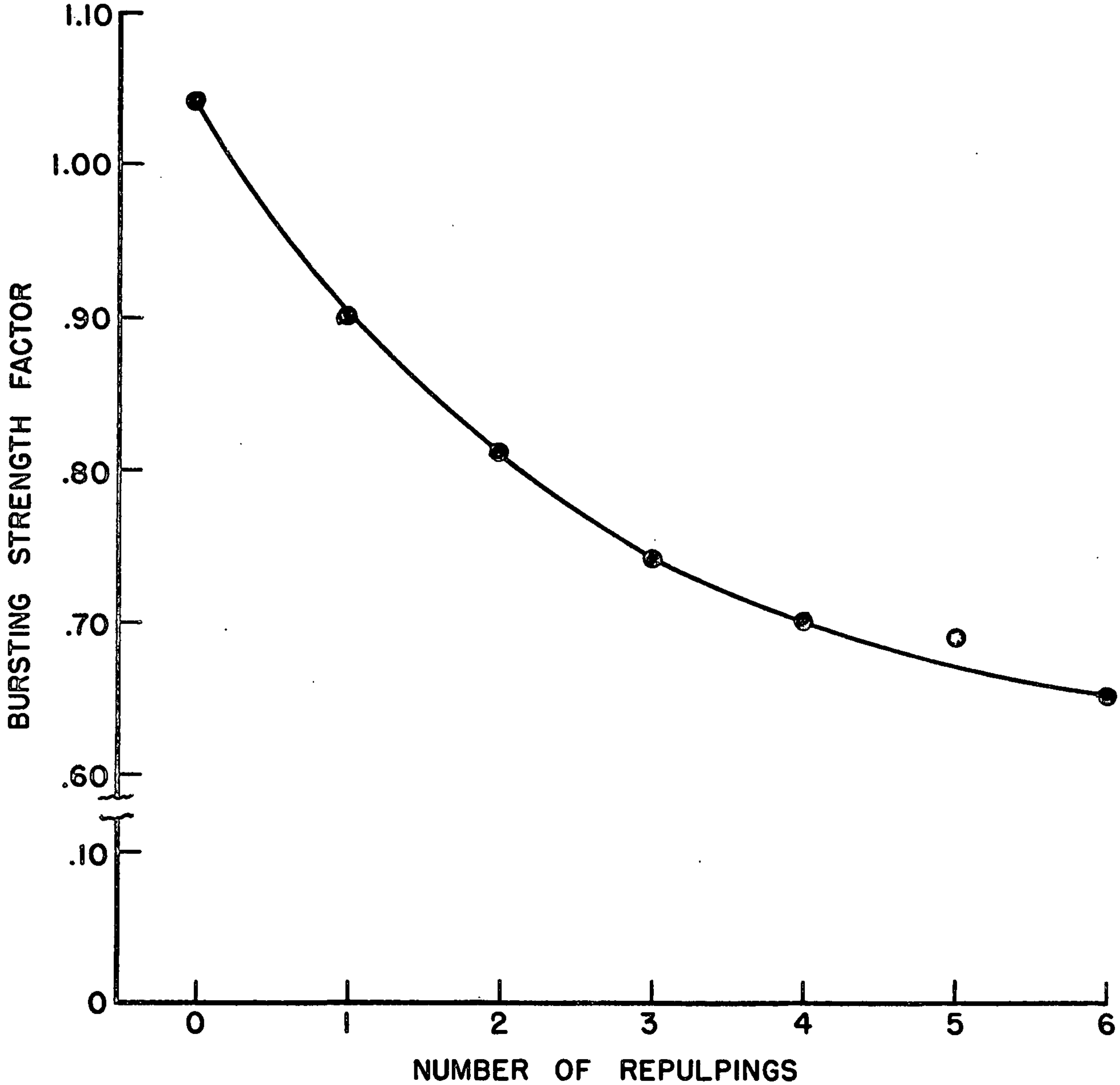

Fig. 14. Effect of Repulping on Bursting Strength
(Open System, Air Dried)

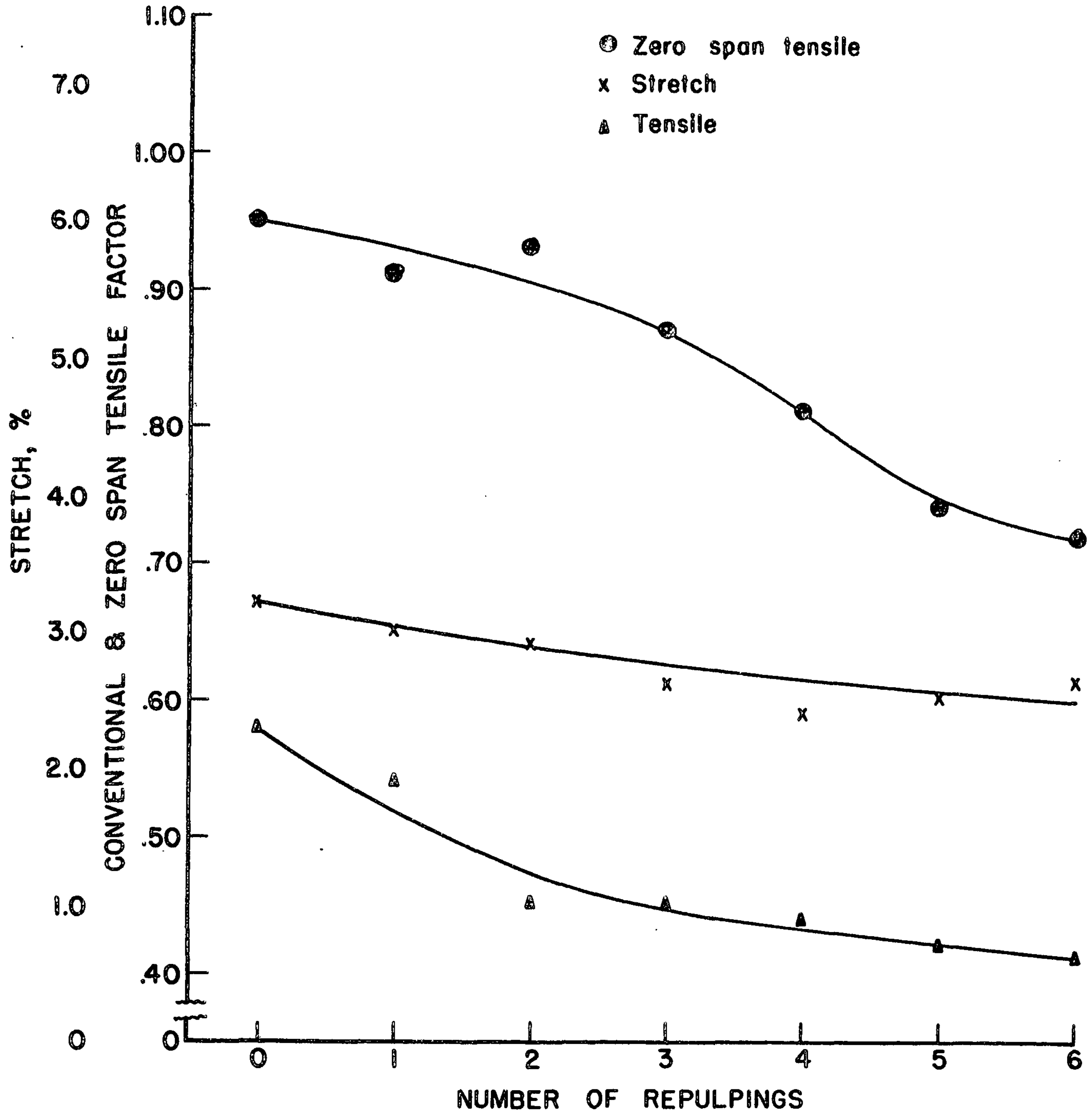

Fig. 15. Effect of Repulping on Tensile and Stretch Characteristics
(Open System, Air Dried)

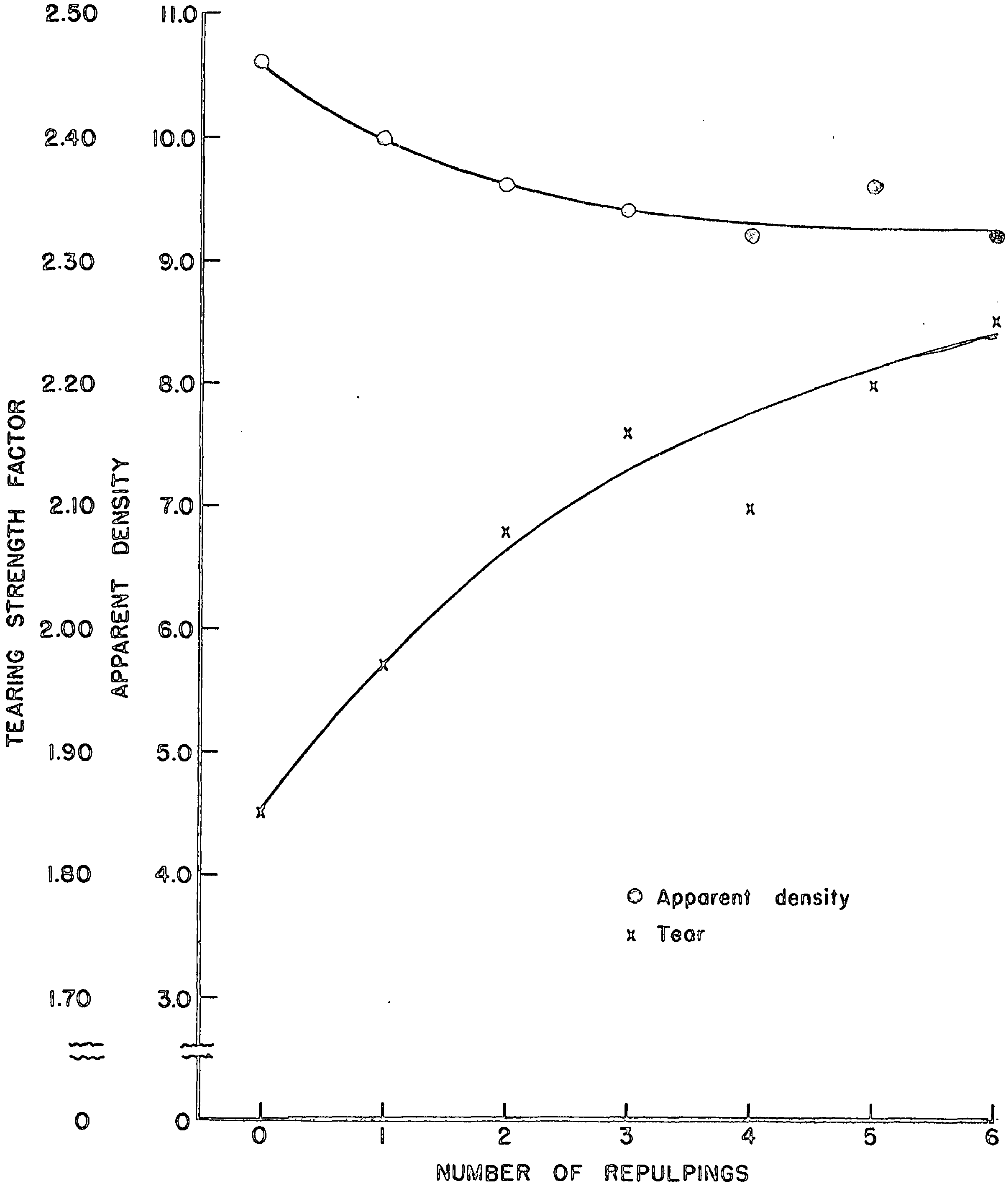

Fig. 16. Effect of Repulping on Apparent Density and Tearing Strength
(Open System, Air Dried)

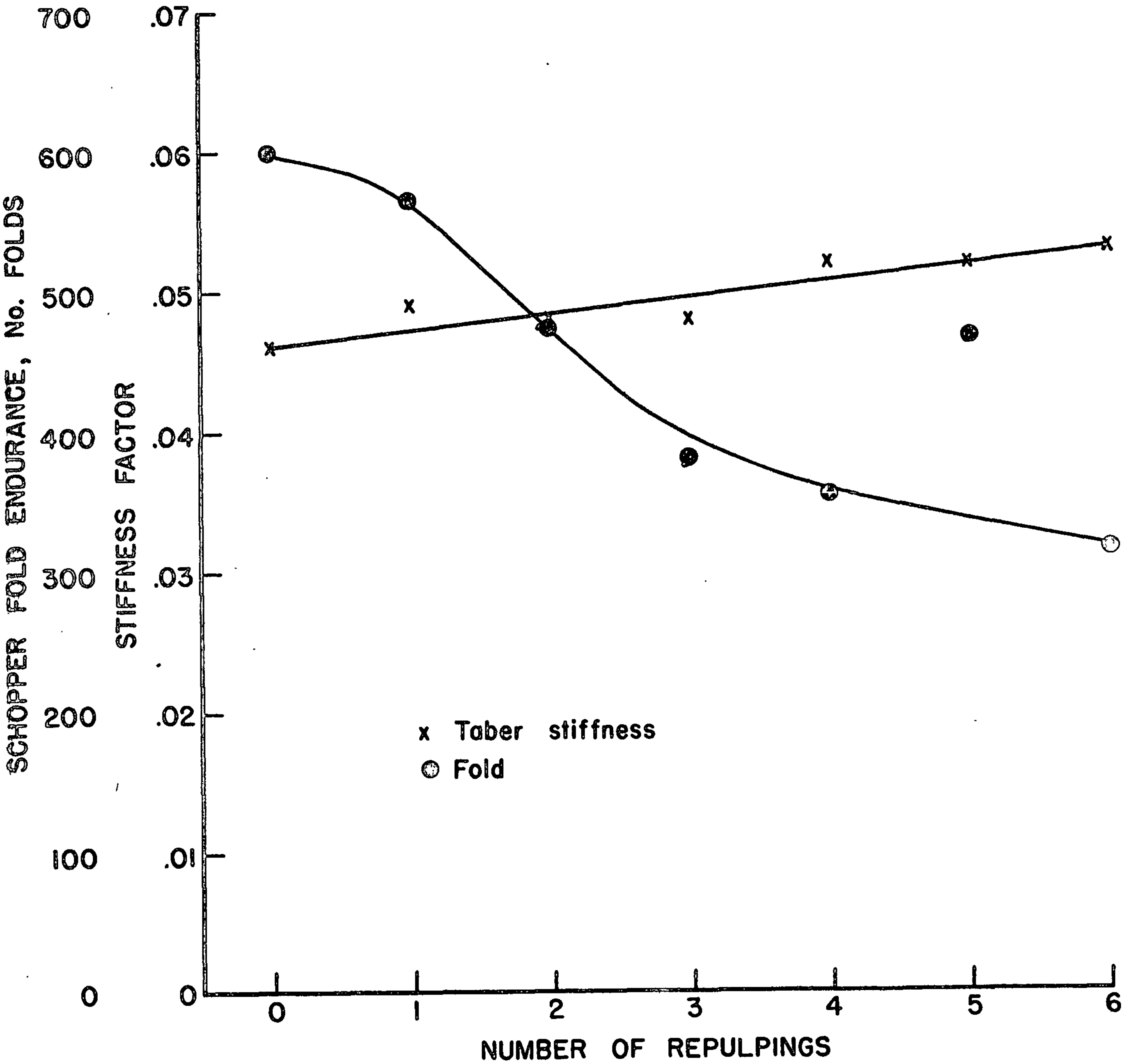

Fig. 17. Effect of Repulping on Fold Endurance and Taber Stiffness
(Open System, Air Dried)

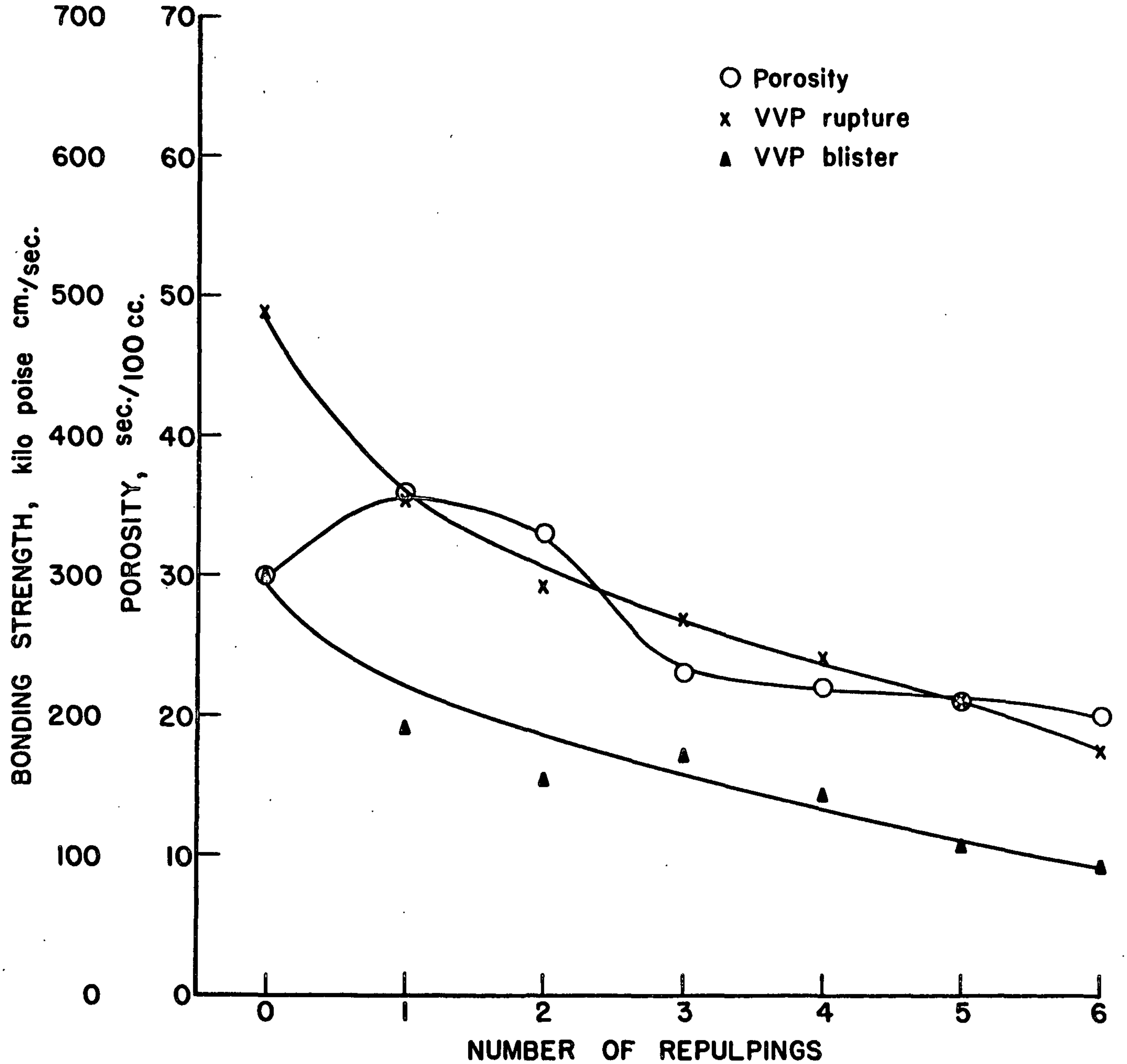

Fig. 18. Effect of Repulping on Porosity and Bonding Strength
(Open System, Air Dried)

lower than initially. The greatest rate of change was in the first two cycles. The magnitude of the change was slightly greater than was obtained for the sheets made with a closed white water system and drum drying. The zero-span tensile exhibited only a slight decrease in the first and second repulping but a progressive decrease thereafter with number of times repulped, being approximately 24% lower after the sixth repulping than initially. The behavior at the 3 to 6 repulpings is quite a departure from that observed for the sheets made with a closed white water system, although the behavior at the first and second is confirmatory. It would appear questionable that the differences reflect solely the difference in forming conditions. The results of the stretch test plotted in Figure 15 indicate that the stretch properties decreased progressively with increase in the number of times repulped. The loss in stretch after six repulpings was approximately 19%.

The relationships between apparent density and tearing strength and the number of times the stock is repulped are shown in Figure 16. As in the case of the handsheets made with the closed system, the apparent density decreased with increase in the number of times the stock was repulped. After the sixth cycle the apparent density was approximately 13% lower than initially. The rate of decrease was greatest for the first and second cycle. The curve for the tearing strength plotted in Figure 16 shows the same trend as was noted for the heat-dried handsheets made with a closed white water system. The greatest rate of increase was for the first and second cycle. The increase after the sixth cycle was approximately 24%. This was slightly less than that observed for the sheets tested in the previous section.

The results of the fold and Taber tests are plotted against the number of times repulped in Figure 17. It may be observed that the Taber stiffness appears to increase slightly with progressive repulpings. It may be recalled that the handsheets made with a closed system did not exhibit any significant change on repulping. Therefore, there is some question as to whether the change observed here is too significant. The fold endurance decreases rapidly with progressive repulpings; at the end of the sixth cycle, the fold endurance was approximately 47% lower than initially.

The relationships between porosity and bonding strength and the number of times the stock is repulped may be seen in Figure 18. The porosity _versus_ repulping curve indicates that the porosity values are quite variant; however, the general trend is for the porosity value to decrease with the number of repulpings indicating that the sheet is less compact and offers less resistance to the passage of air. This behavior is in keeping with the apparent density results. The VVP transverse bonding strength results indicate the same trend as for the sheets made with the closed system, namely, for the bonding strength to decrease sharply with increase in the number of times the stock is repulped. After the sixth cycle the results were approximately 69 and 64% lower for the blister and rupture endpoints than initially. The greatest rates of change occurred on the first repulping. The changes in strength properties with repulping are illustrated in Figures 19 to 22.

In general, the results obtained in this section on handsheets made with an open white water system and air dried exhibited the same trends as the handsheets made with the closed system.

2. Effect of Repulping on Fiber Properties

As previously mentioned, the pulp and various unused stocks were all refined in the same beater using the same procedure to a given freeness level of 615 ± 15 cc. Schopper-Riegler. The stocks used at each level of repulping were evaluated for fiber classification, filtration resistance, and degree of swelling in addition to pH and freeness.

The characteristics of the kraft stock at various repulping cycles are given in Table IV. It may be noted that the freeness of the virgin kraft after soaking in water for four hours and slushing for five minutes was almost 900 cc. After 47 minutes of beating, the average freeness had dropped to 619 cc. On the first repulping cycle using the handsheets prepared from stock refined in the initial cycle, it required only 9.5 minutes of beating time to reach an average

TABLE IV

PROPERTIES OF KRAFT STOCK AT VARIOUS REPULPING CYCLES

No. times repulped	0	1	2	3	4	5	6
Characteristics of stock							
Freeness, cc.							
After 5" slushing	898	728	657	643	658	638	633
End of beating	619	617	613	618	616	612	619
Beating time, min.	4.7	9.5	3.0	2.2	2.5	1.8	1.0
pH							
After 5" slushing	7.97	7.75	7.56	7.75	8.3	8.2	8.5
End of beating	7.91	7.62	7.59	7.68	8.3	8.2	8.5
Degree of swelling, %							
After 5" slushing	108.8	149.4	157.1	150.6	149.0	144.3	137.6
End of beating	219.6	192.9	169.2	159.3	158.6	153.5	141.0
Filtration resistance[1], $R \times 10^{-8}$ cm./g.							
End of beating							
Frictional pressure drop, cm. H_2O							
10	3.29	4.30	4.82	4.23	4.66	4.65	4.74
20	5.02	6.55	7.38	6.40	7.06	7.09	7.22
30	6.49	8.53	9.58	8.22	9.13	9.20	9.28
40	7.85	10.29	11.54	9.85	10.96	11.06	11.14
50	9.12	11.95	13.35	11.40	12.69	12.82	12.84
60	10.34	13.52	15.11	12.85	14.30	14.45	14.48
70	11.53	15.01	16.80	14.25	15.88	16.04	16.05
80	12.66	16.52	18.44	15.55	17.32	17.56	17.50
90	13.76	17.89	20.01	16.85	18.80	19.01	18.98

TABLE IV (Continued)

PROPERTIES OF KRAFT STOCK AT VARIOUS REPULPING CYCLES

No. times repulped	0	1	2	3	4	5	6
Filtration resistance (Continued)							
After 5 min. slushing							
Frictional pressure drop, cm. H_2O							
10	0.265						
20	0.362						
30	0.440						
40	0.507						
50	0.569						
60	0.625						
70	0.680						
80	0.731						
90	0.782						
Effective specific volume, $cm.^3/g.$	1.30	1.35	1.70	1.45	0.97	1.23	--
Effective specific surface[2], $cm.^2/g.$	41,400	43,400	44,200	40,000	47,800	42,900	--
Compressibility, g./cc.							
Pad concentration end of beating at							
Compact pressure, cm. H_2O							
17.3	0.0885	0.0985	0.096	0.102	0.103	0.116	0.118
26.3	0.110	0.121	0.116	0.122	0.131	0.141	0.140
40.1	0.138	0.149	0.137	0.146	0.152	0.178	0.170
62.3	0.175	0.182	0.168	0.180	0.182	0.206	0.209
100.0	0.223	0.228	0.204	0.220	0.235	0.243	0.260

TABLE IV (Continued)

PROPERTIES OF KRAFT STOCK AT VARIOUS REPULPING CYCLES

No. times repulped	0	1	2	3	4	5	6
Compressibility, g./cc. (Continued)							
After 5 min. slushing							
Compact pressure, cm. H_2O							
17.3	0.0839						
26.3	0.0974						
40.1	0.112						
62.3	0.138						
100.0	0.1625						
Fiber classification, %							
End of beating							
On 20-mesh	76.1	74.3	75.4	72.0	72.7	74.1	71.7
On 35-mesh	4.7	4.0	5.3	5.6	5.0	4.4	5.7
On 65-mesh	4.4	4.5	5.4	5.8	6.4	6.8	4.3
On 150-mesh	3.2	3.1	4.0	4.9	4.1	4.1	4.3
Through 150-mesh[3]	11.6	14.2	10.0	16.8	12.0	10.6	11.3
After 5" slushing							
On 20-mesh	89.5						
On 35-mesh	3.3						
On 65-mesh	2.3						
On 150-mesh	1.0						
Through 150-mesh	4.0						

Notes:
1. Average of two or more determinations.
2. Specific surface for pulp after 5 min. slushing was 8,500 $cm.^2/g.$
3. Obtained by difference

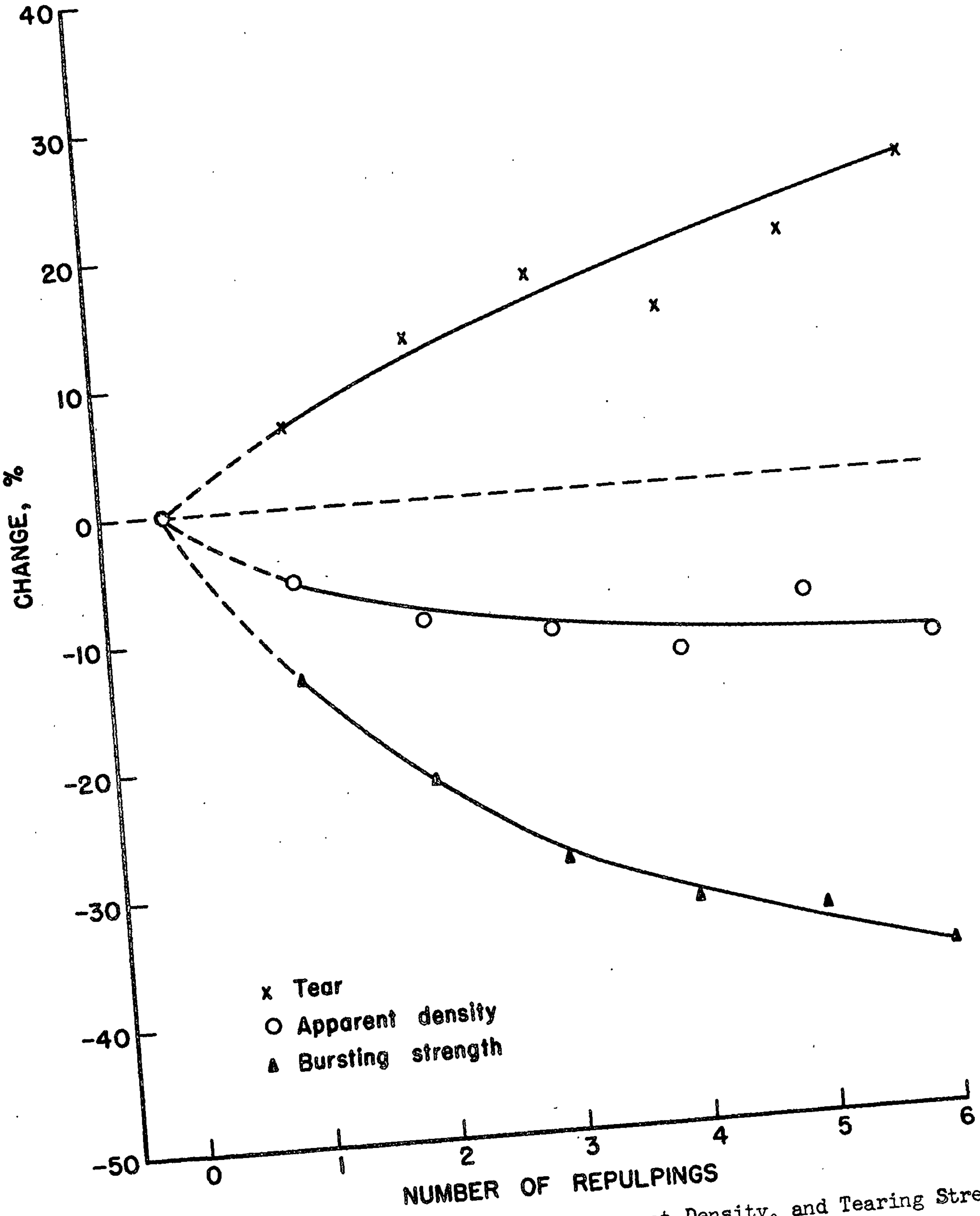

Fig. 19. Change in Bursting Strength, Apparent Density, and Tearing Strength Sith Repulping
(Open System, Heat Dried)

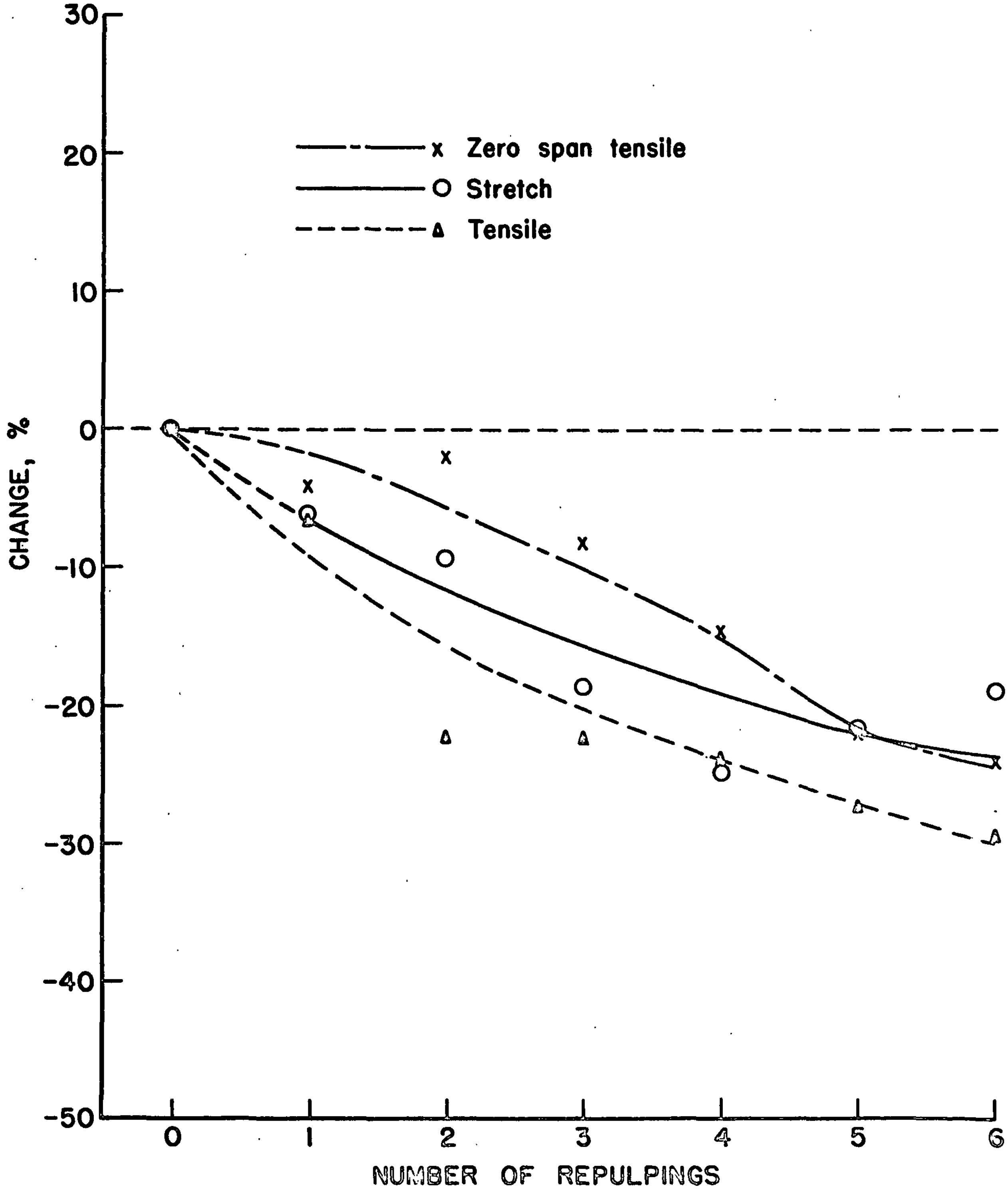

Fig. 20. Change in Tensile and Stretch Characteristics with Repulping
(Open System, Air Dried)

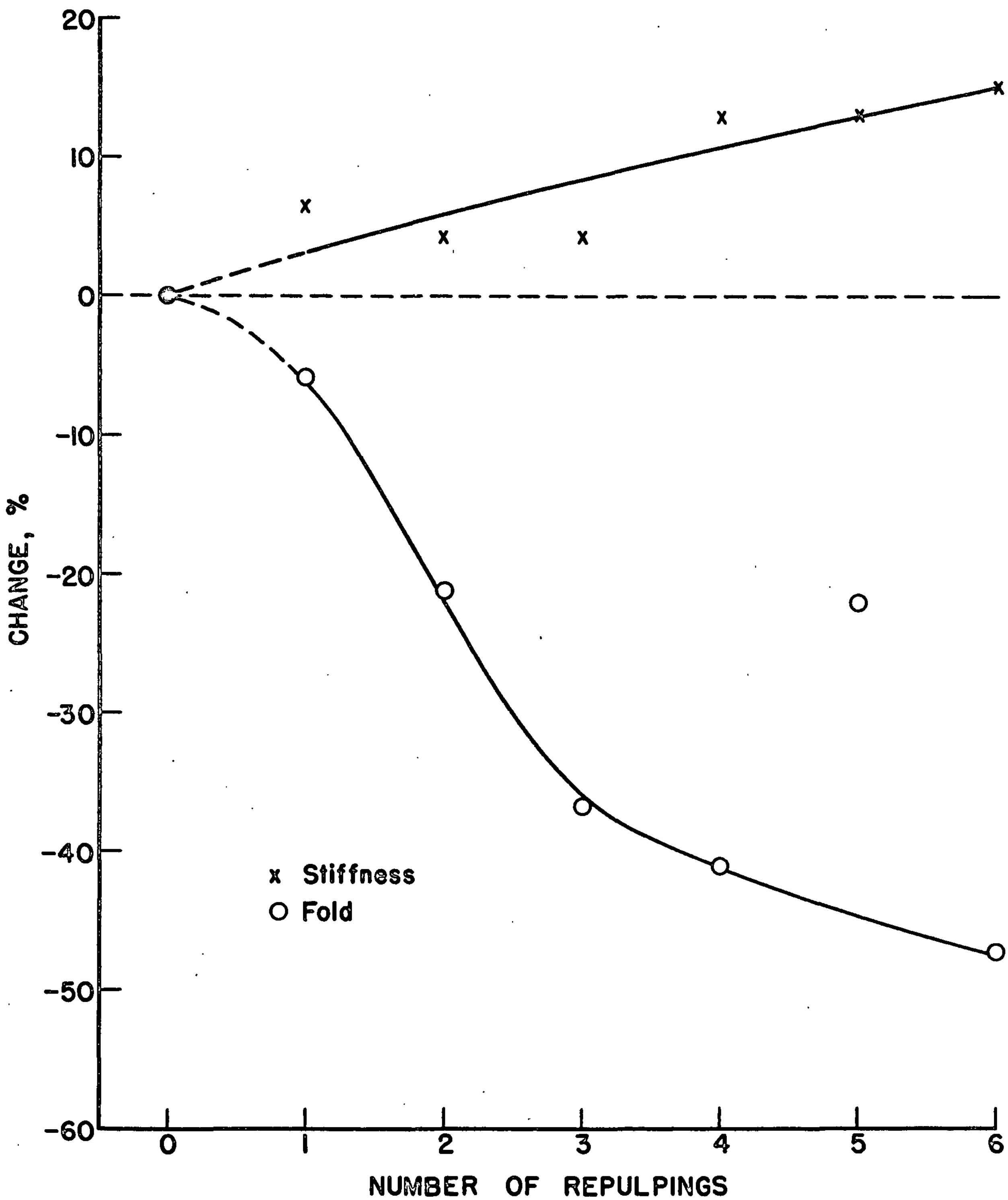

Fig. 21. Change in Fold Endurance and Taber Stiffness with Repulping
(Open System, Air Dried)

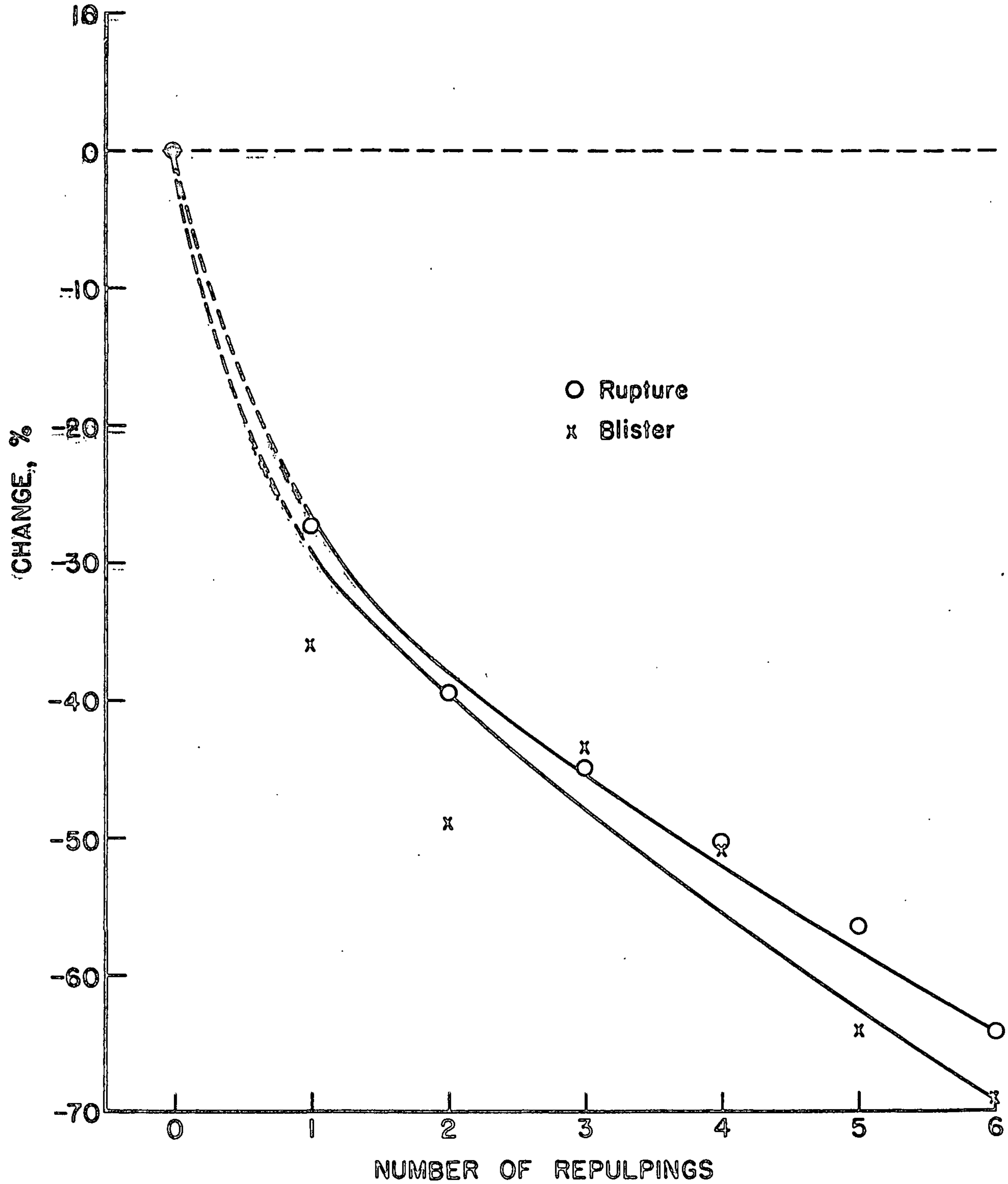

Fig. 22. Change in Bonding Strength with Repulping
(Open System, Air Dried)

freeness level of 617 cc. Thereafter, one to three minutes of beating preceded by five minutes of slushing was sufficient to reduce the freeness to the desired level of 615 ± 15 cc.

The pH of the stock in the beater varied throughout the entire study from a low of 7.56 to a high of 8.5. Inasmuch as no size, alum or other beater additions were added to the stock, the change in pH probably is representative of the variation in the tap water which was used in all beater runs.

The changes in fiber length with the initial beating or refining cycle and subsequent repulping cycles may be noted from the data tabulated in Table IV and graphically presented in Figures 23, 24, and 25. It may be seen from the bar chart presented in Figure 23 that after only 5 minutes slushing 4% of the pulp was of such a size that it passed through a 150-mesh screen. As may be noted, the 47 minutes of beating in the initial cycle resulted in a decrease of 13.4% in the stock retained on the 20-mesh. This reduction of the long fibers appears to have been distributed throughout the other fractions with Through-150 receiving the largest percentage. The actual changes were as follows:

On-20--13.4 points reduction in per cent retention
On-35-- 1.4 points increased in per cent retention
On-65-- 2.1 points increased in per cent retention
On-150-- 2.2 points increased in per cent retention
Through-150-- 7.6 points increased in per cent retention

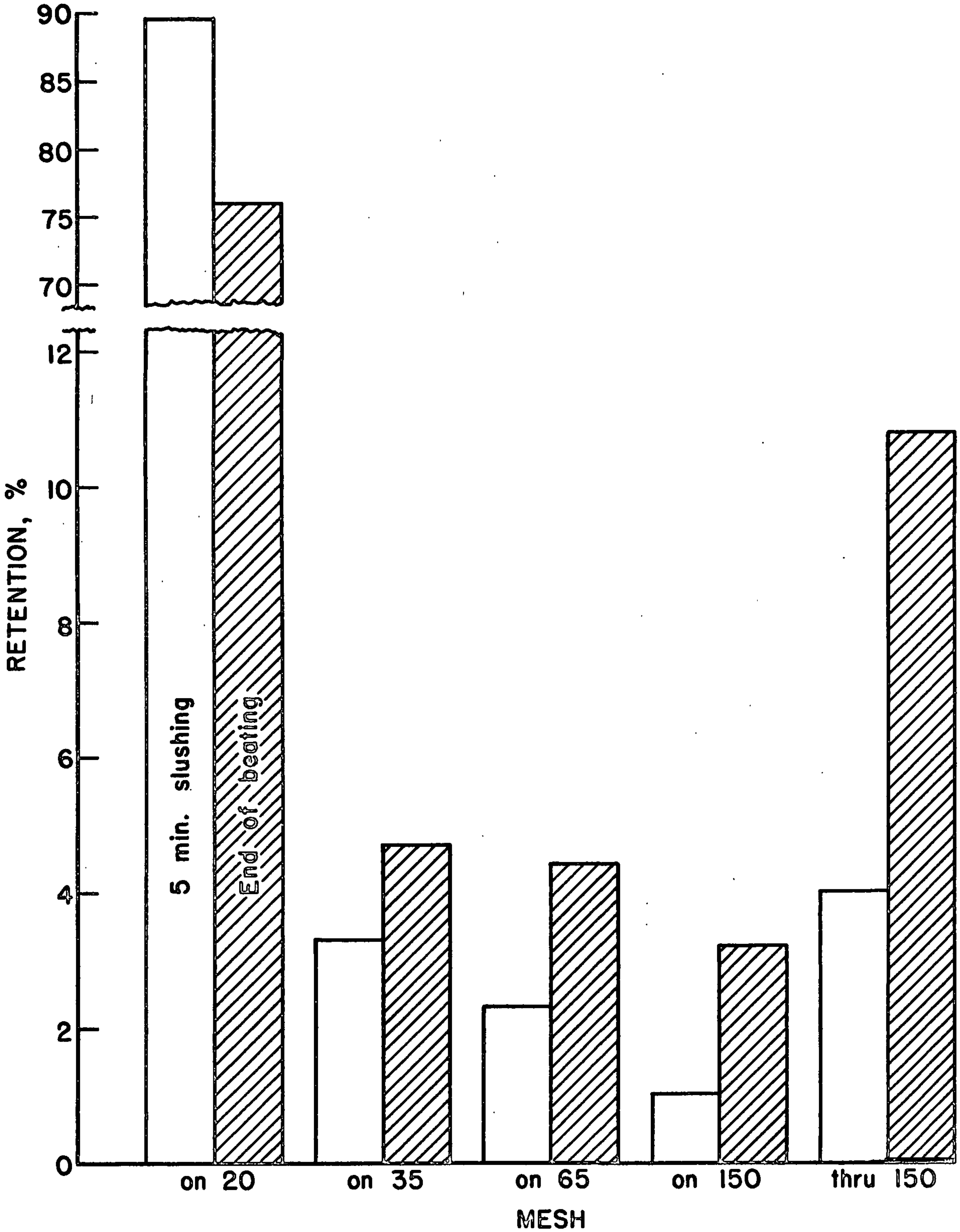

Fig. 23. Fiber Length Classification Before and After Beating
(Initial Cycle)

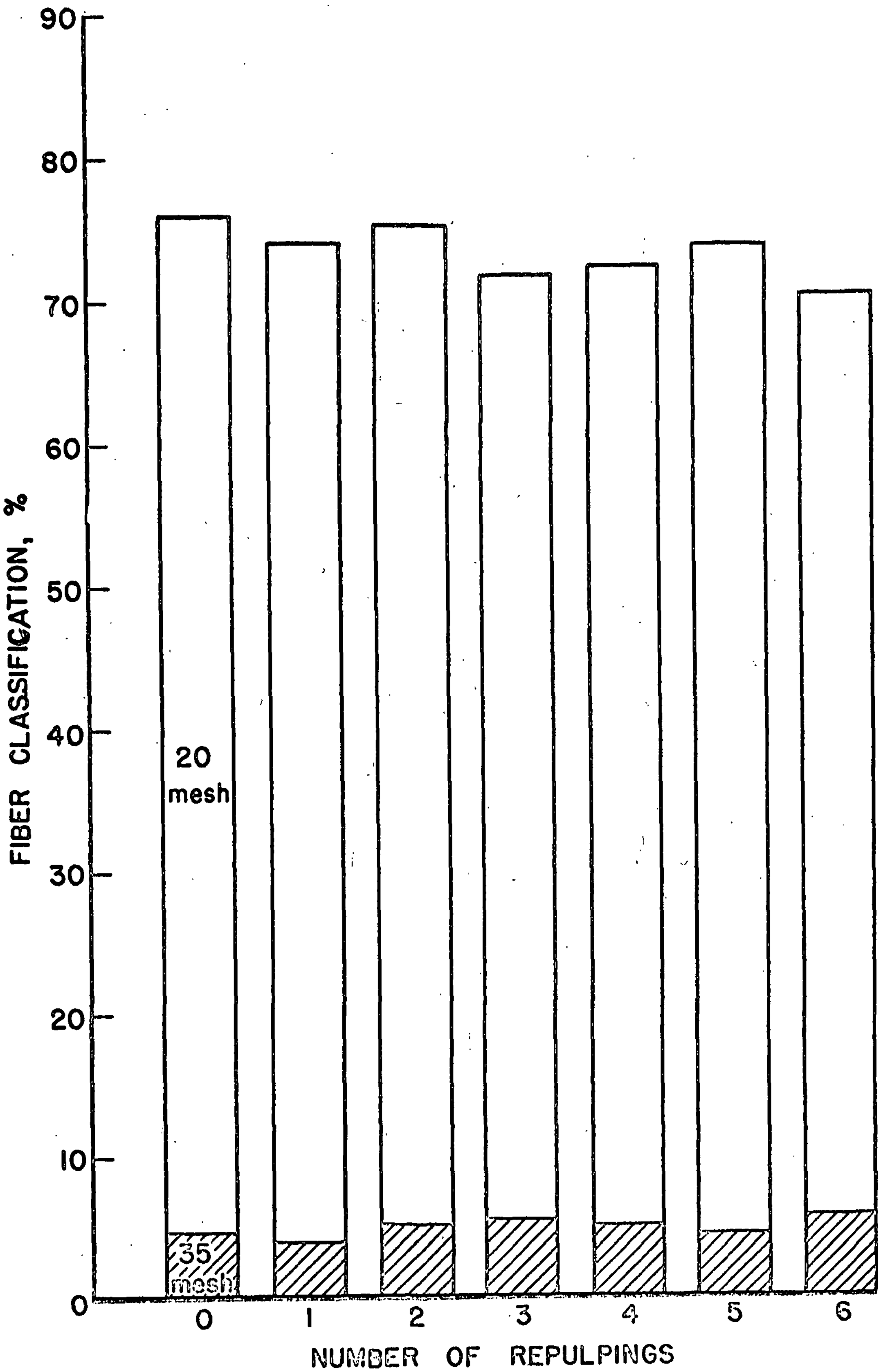

Fig. 24. Effect of Repulping on Fiber Classification

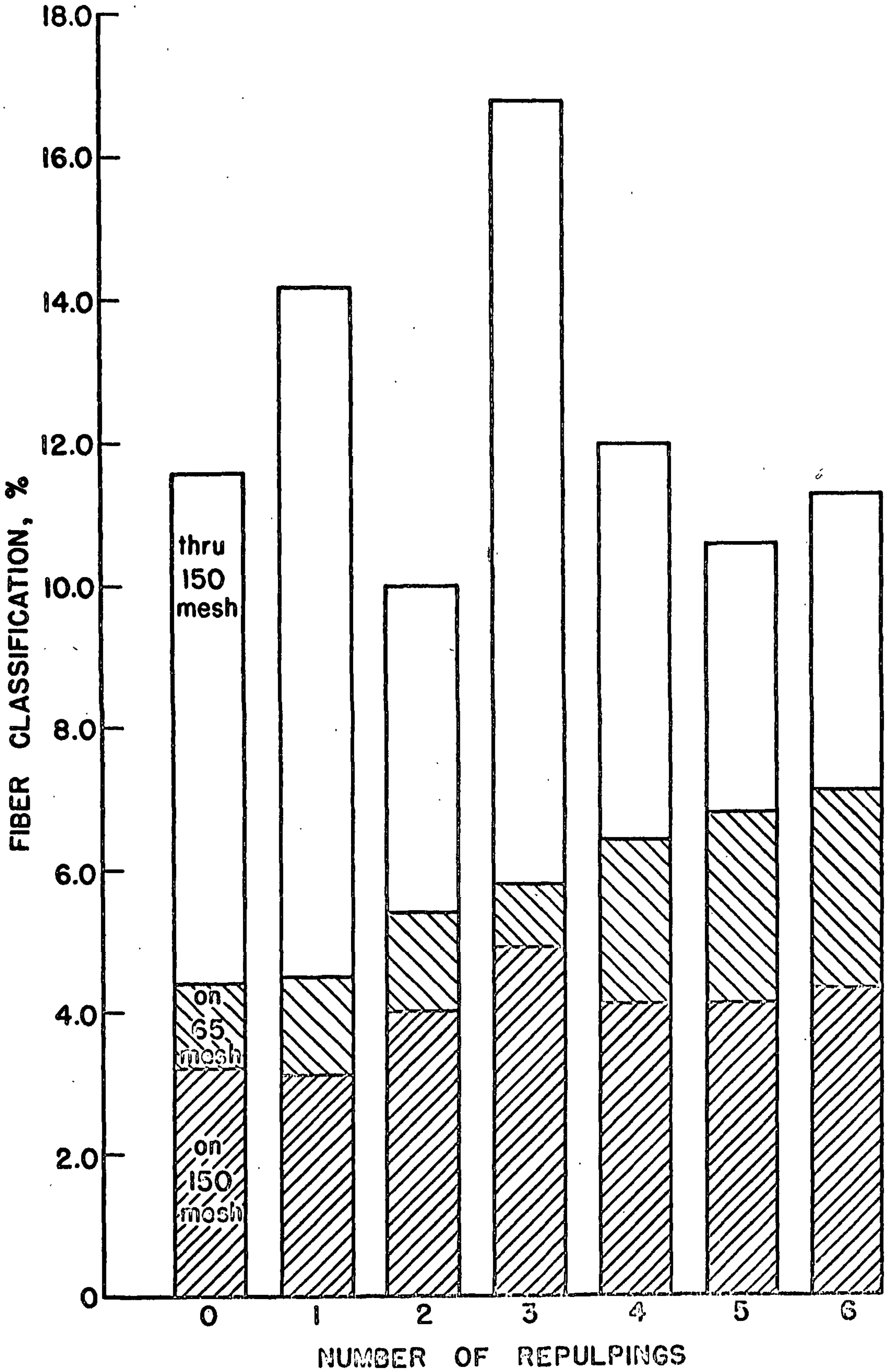

Fig. 25. Effect of Repulping on Fiber Classification

On the basis of the above results it appears that in so far as fiber length is concerned, the main effect of the beating in the initial cycle was a decrease in the On-20 fraction and an increase in the Through-150 fraction. The increases at the other screen-mesh levels were only slightly greater than the precision of the test, namely, on duplicate runs on the same sample the results at each level should agree with 1% retention--i.e., 1.0-2.0 and 75.0-76.0.

A comparison of the change in fiber length on progressive repulpings and beating to the same freeness level may be seen from the data plotted in Figures 24 and 25. It may be seen that the On-20 mesh fraction veried from a high of 76.1% retention for the initial cycle to 71.7% retention for the sixth repulping cycle. The per cent retentions On-20-mesh for the initial and six repulping cycles are tabulated below.

Cycle	Retention, %
0	76.1
1	74.3
2	75.4
3	72.0
4	72.7
5	74.1
6	71.7

With the exception of Repulping cycle 5, there is a tendency for the amount retained on the On-20-mesh to decrease after the second repulping cycle. It is questionable, however, whether the decrease is significant inasmuch as the values for all the cycles are dispersed within

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Due to a typographical error, there are no pages 101 and 102.

a range of ± 2.2 of the average of 73.8%. The results obtained for the On-35-mesh fraction (see Figure 24) indicate that the results are dispersed within a range of approximately ± 0.9 of the average of 4.9%. Although the significance of the differences is open to question, there appears to be a slight tendency for the amount retained to increase after about the second cycle. It may be recalled that this tendency is just the opposite of what was observed for the On-20-mesh fractions. The amounts retained on the On-35-mesh for the various repulping cycles are given below:

Cycle	Retention, %
0	4.7
1	4.0
2	5.3
3	5.6
4	5.0
5	4.4
6	5.7

The amounts retained on the On-65 and On-150-mesh are tabulated below and graphically illustrated in Figure 25.

	Retention, %	
Cycle	On-65	On-150
0	4.4	3.2
1	4.5	3.1
2	5.4	4.0
3	5.8	4.9
4	6.4	4.1
5	6.8	4.1
6	7.1	4.3

It may be noted from the data plotted in Figure 25 for the On-65-mesh that there seems to be a very small but progressive increase in the amount of fiber retained on the On-65-mesh with increase in the number of times the fiber is repulped. In the case of the On-150-mesh there appears to be an increase after the first repulping; however, it is questionable if the change is significant.

The Through-150 fractions are tabulated below and also graphically illustrated in Figure 25.

Cycle	Retention, %
0	11.6
1	14.2
2	10.0
3	16.8
4	12.0
5	10.6
6	11.3

It should be mentioned that the Through-150 values were not measured values but calculated values obtained by difference; thus, they reflect to a degree the variability associated with the determination of the other fractions. With the exception of the values for Cycles 1 and 3, there would not appear to be a significant change with repulping.

When all the fractions are considered, it may be seen that there was very little change in fiber length introduced as a result of progressive repulpings to a given freeness level. The appearance of the stock at the end of beating for Cycles 0 and 6 as may be seen from Figures 26 and 27 tend to confirm the above. In practice, the waste stock, particularly if it comes in as corrugated board and boxes, was

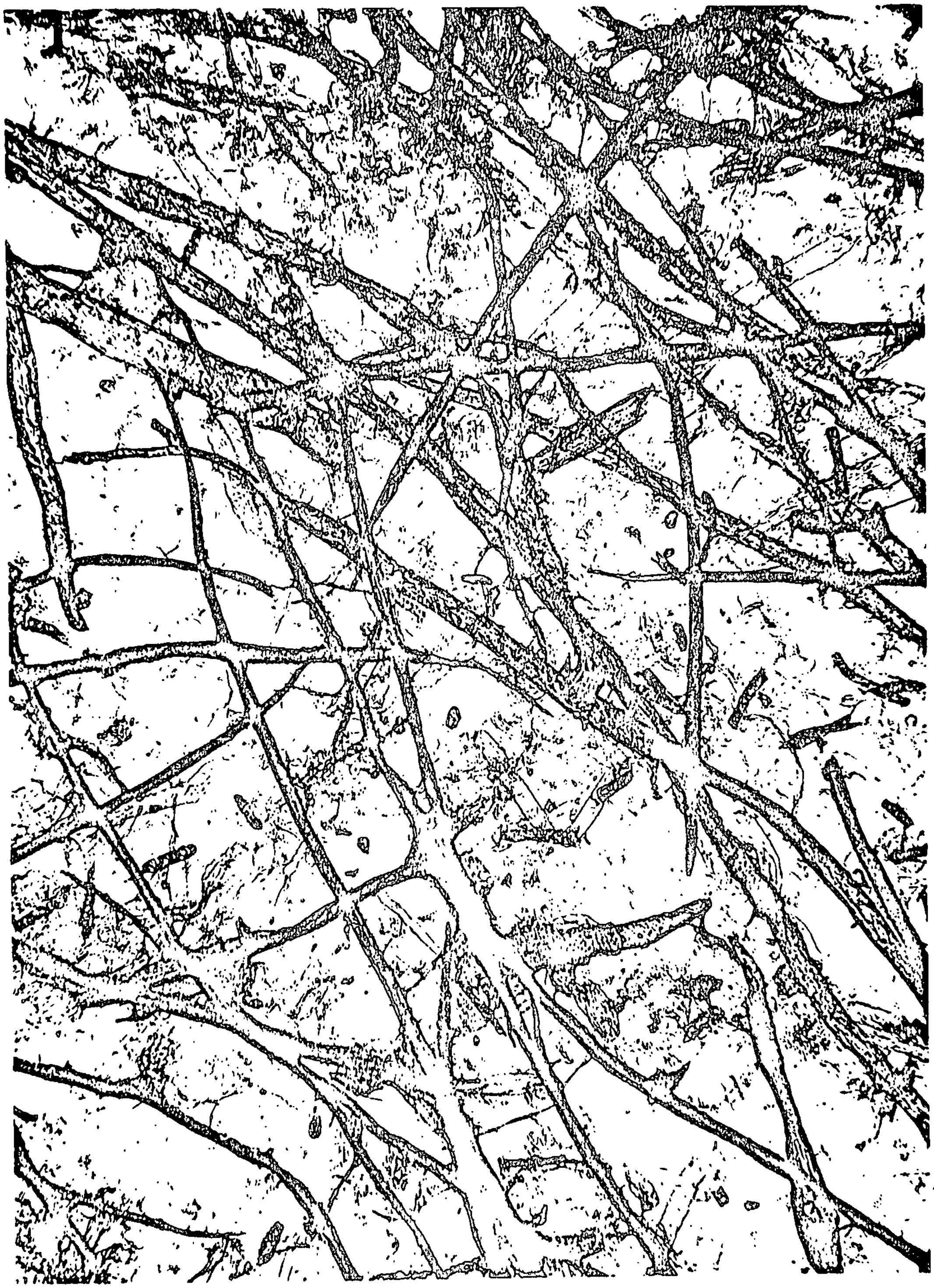

Fig. 26. Photomicrograph of Stock at End of Beating, Cycle 0

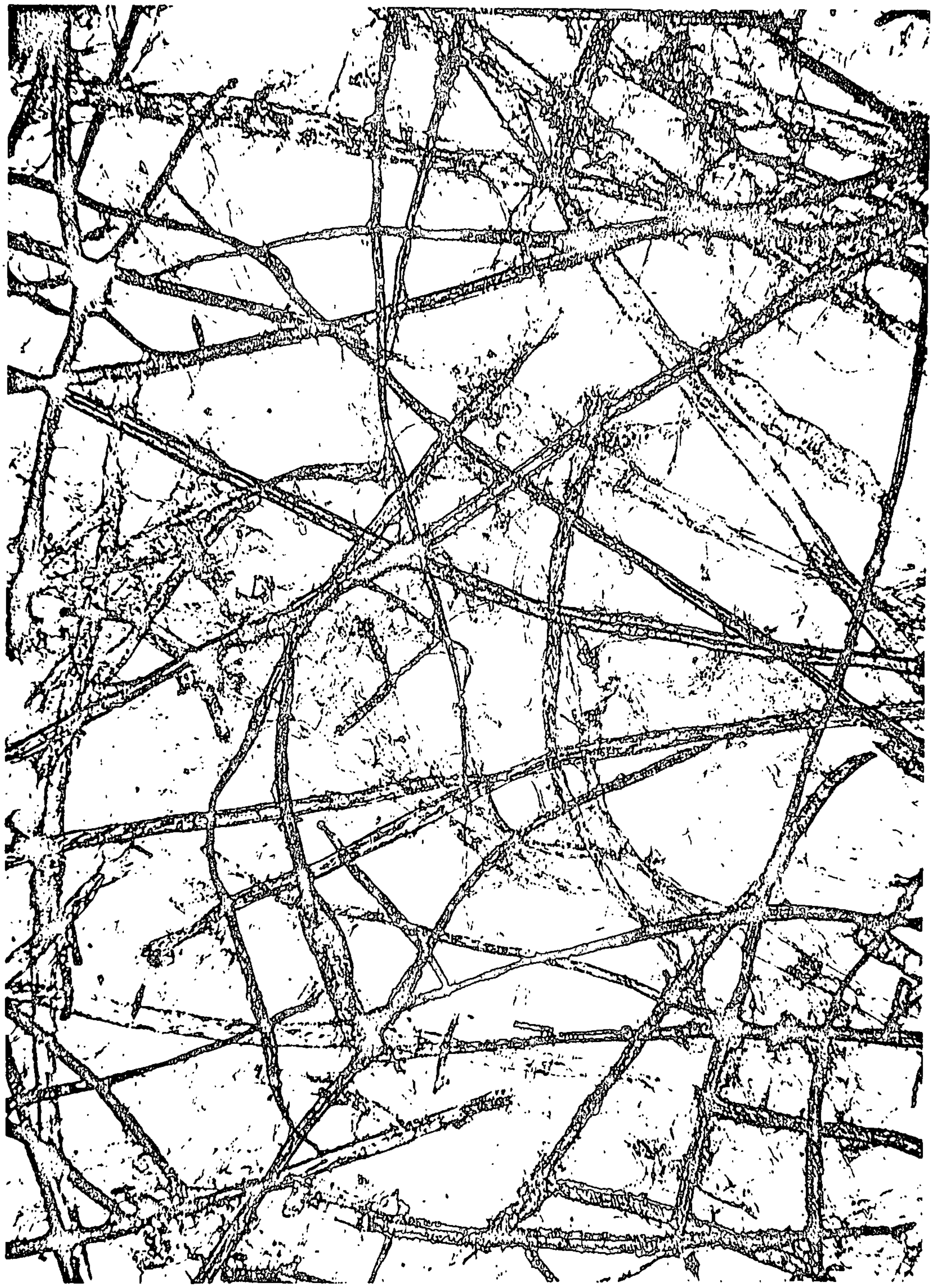

Fig. 27. Photomicrograph of Stock at End of Beating, Cycle 6.

not refined to as low a freeness when made into Fourdrinier board initially as it is when repulped in the manufacture of jute liner board. Thus, in practice, one would expect not only a drop in freeness but also a change in fiber-length classification. The purpose in maintaining a constant level of freeness was to see if the stock when so refined would exhibit changes in fiber length and if no significant change occurred to determine the extent to which sheet properties changed in the absence of changes in fiber length.

As mentioned previously, filtration resistance measurements were made on the beaten stock at each cycle of repulping. The filtration resistance is of value in predicting drainage rates and is in somewhat the same category as freeness from which one too frequently tries to predict the resulting sheet strength. Studies carried out, however, have shown that under a given set of conditions there is a direct relationship between filtration resistance and the rate of water removal. The filtration resistance per se provides information only on rate of water removal from the stock and does not provide any measure of the strength characteristics. Recent studies have indicated, however, that when the filtration resistance is broken down into its component--i.e., specific volume, specific surface and compressibility--considerably more insight is provided relative to the potential strengths of the stocks. The filtration resistance, from which the components are obtained, does not provide any insight as to strength because two given pulps may have the same filtration resistance--i.e., drainage rate would be the same under like conditions of pressure, etc., but because of the interplay of the components of filtration resistance, these two pulps may have entirely different properties. The basic equation for filtration resistance, R, is:

$$R = \frac{k \quad S^2 \Delta P_f}{\int_0^{\Delta P_f} \frac{(1-vc)^3}{c} \, d\,(p)} \qquad (2)$$

where k = Kozeny constant

S = specific surface (fiber)

ΔP_f = pressure drop across fibrous pad

v = the effective specific volume

p = mechanical compacting pressure developed at a point in the bed

c = the point concentration of bed material at a differential layer in the filter mat where compacting pressure is p.

The complete filtration resistance data are tabulated in Table IV and illustrated in Figures 28 and 29.

The results tabulated in Table IV are the average test results obtained for each cycle. The average deviation of filtration resistance from the mean of all the beater runs tested at each cycle of repulping may be seen from the following:

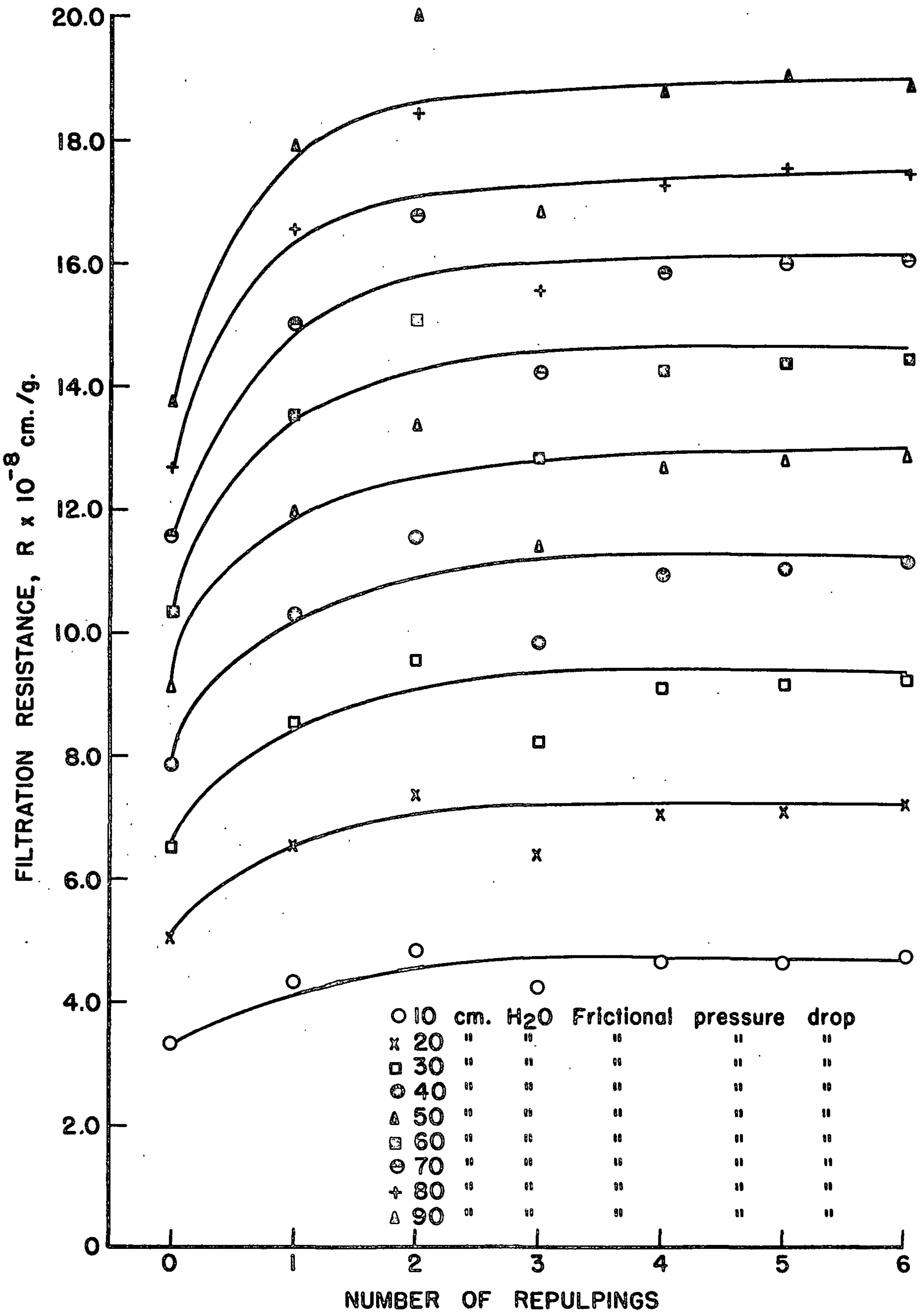

Fig. 28. Effect of Repulping on Filtration Resistance

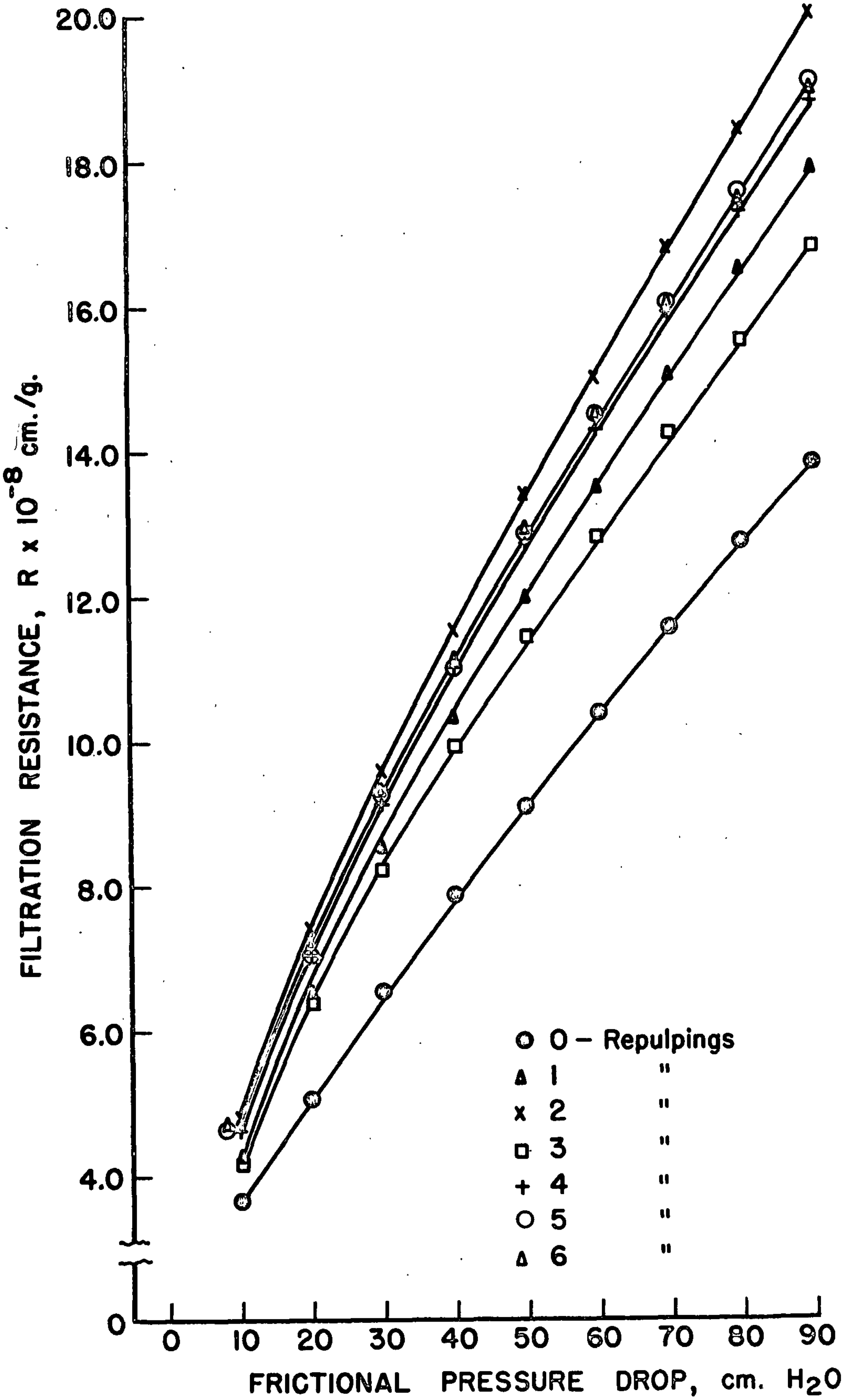

Fig. 29. Effect of Repulping on Filtration Resistance

Cycle	Average Deviation, %
0	2.5
1	2.5
2	0.3
3	2.0
4	2.4
5	3.3
6	2.2

The above indicates that the reproducibility of beater conditions within a given cycle, as reflected by filtration resistance, was fairly good. It was observed, however, that when duplicate tests were made on stock from a given beater, the results agreed within about 0.5%; thus, the agreement within a given beater was approximately 0.5% compared to approximately 2.5% between beaters.

The data in Figure 28 illustrates the relationship between filtration resistance and the number of times repulped for various levels of frictional pressure drop--i.e., pressure drop across filter mat which in this case was composed of the stocks in question. Figure 29 is a replot showing the effect of increased frictional pressure drop on the filtration resistance of a given stock. It may be observed that at all levels of frictional pressure drop, the filtration resistance increased on repulping. The most significant change came in the first repulping wherein the filtration resistance increased approximately 30% and then appeared to level off as the increase between the first and sixth repulping was only about 7.0%. The index used to give each repulping cycle the same degree of beating was freeness, a value of 615 ± 15 cc. S.-R. being the chosen point. The data clearly indicate that refining to the same freeness value does not assure equality of filtration resistance or strength properties. An increase in the filtration resistance of the order of magnitude shown here would be reflected in slower drainage on the paper machine. The increase in filtration resistance does not appear to be strongly related to fiber length as there was only a small change in the fractionation tests on repulping; thus, it would appear that neither freeness nor fiber length are a criterion on which to base drainage characteristics.

It was mentioned earlier that the components of filtration resistance are related to strength properties. The three components of filtration resistance are (a) specific surface, (b) effective specific volume, and (c) compressibility. Specific surface is the total external surface area of the fibers. It is the surface area which the water "sees" or contacts as it flows through a pad of fibers. The effective specific volume is that volume per unit mass of fibers which is denied to flow or which resists flow--the external volume of the fiber plus the volume occupied by any immobilized or bound water on the surface of the fibers. Thus, it is related to the degree of swelling. The compressibility is the apparent bed density of the wet pad at a given compacting pressure. Although much work remains to be done to thoroughly explore the relation between the components of filtration resistance and strength properties, preliminary investigations indicate that this is a very promising approach to pulp and stock evaluation and should contribute much to the technology of strength development. The investigations to date indicate certain generalizations which for the time being appear to be in order. These are: the greater the specific surface, the greater the potential area for bonding. Normally, the greater the specific surface the greater the strength. Since the specific surface, S, is in the numerator of Equation (2), the higher the specific surface the greater the filtration resistance and the more difficult the stock is to drain. As a first approximation, the filtration resistance increases as the square of the specific surface. Because of their position in the equation for filtration resistance, specific volume and compressibility should have only minor effects on filtration resistance in contrast to specific surface. Other things being equal, for the same specific surface the larger the compressibility and/or specific volume, the greater the anticipated filtration resistance.

Little is known as to the relationship of compressibility to strength. The main reason for determining it in this study is that it is necessary to the determination of specific surface and specific volume. It may be fair to postulate that the greater the compressibility the greater the flexibility of the fibers. The specific volume is related to specific surface; however, it is possible to have stocks of the same specific volume, but differing as to specific surface due to fiber geometry. Although specific surface is related to specific volume, one may be changed without introducing a change in the other; e.g., it has been found that if a given pulp is fractionated, each fraction will have a different specific surface, but the specific volume will be the same. In any normal beating process the specific surface and the specific volume increases. Compressibility does not increase if the beating is mostly cutting.

The results obtained for specific surface are tabulated in Table IV. Surprisingly, the external surface area of the stocks did not appear to change significantly with repulping. The variability of the specific surface data about the average of 43,300 square centimeters per gram for the beaten stock is not considered significant. This would appear to be in keeping with the fiber fractionation data.

The compressibility values as determined experimentally for each beating cycle are tabulated in Table IV and illustrated in Figure 30. As may be seen from Figure 30, pad concentration or compressibility increases with repulping. Because of the use of very thin pads, necessitated by the high filtration resistance, the compressibility measurements suffered from a lack of precision. Error introduced from this source has a negligible effect on the calculated value of specific surface but has a marked effect on the calculated specific volumes. Thus, because of the apparent poor precision of the compressibility measurements, the specific volume data in Table IV shows no clearly defined trend with repulping. Inasmuch as the specific volume is the

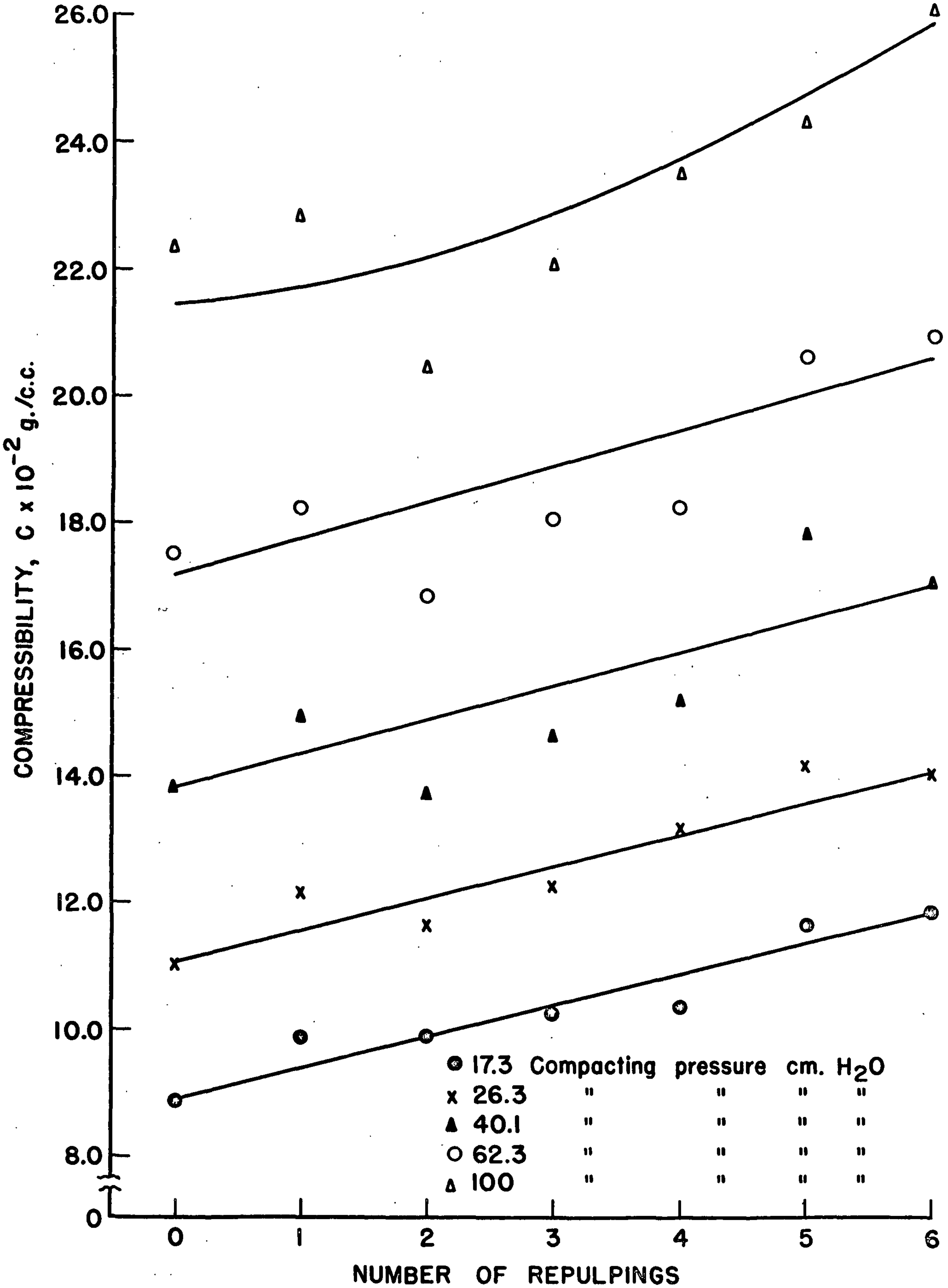

Fig. 30. Effect of Repulping on Compressibility Characteristics of Stock

hydrodynamic volume which resists the flow of water through the pad, it is related to the degree of swelling and one would anticipate that it will correlate well with the degree of swelling. Unfortunately, because of the above-mentioned error, the specific volume data obtained in this study is of no value.

As was mentioned earlier, papermaking fibers have the characteristic of swelling when in contact with liquid water. The swelling is the result of the affinity of the fibers for water. As the fiber takes up water it swells thereby increasing in softness, flexibility and potential bonding because of greater area and more intimate contact of the fibers on drying. The results of the degree of swelling as measured by the Jayme method at each repulping cycle are given in Table IV and illustrated in Figure 31. It may be seen that the degree of swelling decreases with repulping, the most rapid decrease being in the first and second cycle. After 5 minutes slushing in the beater, the virgin stock had a swelling value of 108.8%. After 47 minutes of beating in the Valley beater, the swelling value was 219.6%. The first time the handsheets were repulped, the swelling was 149.4 and 193.9% for 5 minutes slushing and at the end of the beating cycle, respectively. At the end of the above beating periods, the stocks had the same freeness (615 ± 15 cc. S.-R.) but the swelling had decreased approximately 12% on repulping.

Several seemingly important conclusions may be drawn from a

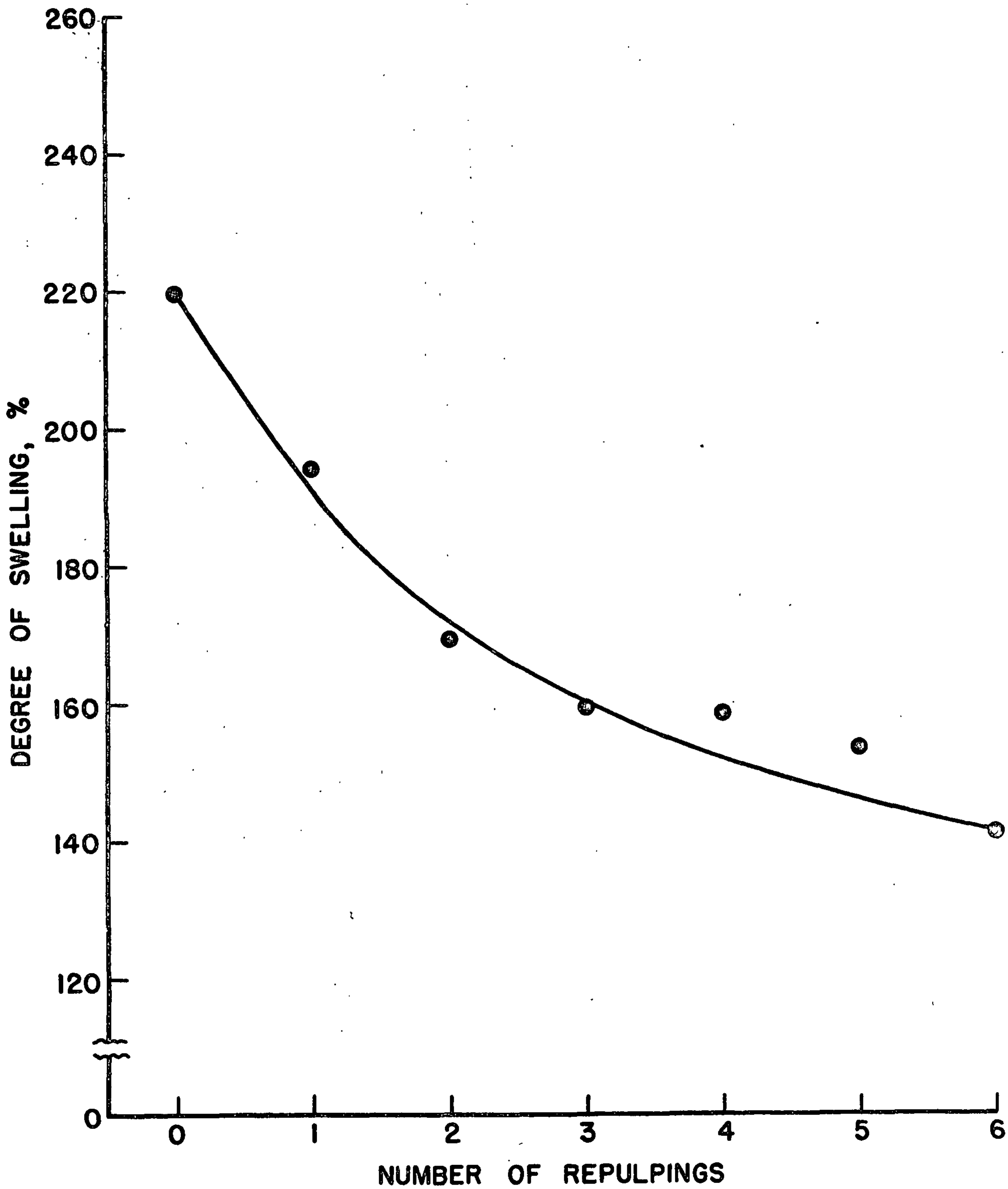

Fig. 31. Effect of Repulping on Degree of Swelling

consideration of the observed behavior of the repulped stocks and the properties of the handsheets made therefrom. The constancy of the specific surface as measured in the filtration test is confirmed by the Bauer-McNett fiber classification data (see Table IV and Figures 23, 24, 25) wherein it may be seen that neither the long fibers nor the fines (Through-150) changed significantly with repulping.

Since the fiber length distribution was not altered significantly, the observed increase in wet pulp pad compressibility (see Figure 30) may have been the result of an increase in wet-fiber flexibility. One might postulate that as repulping progresses the fiber structure is degraded and weakened, with a resulting increase in flexibility. The results of the zero span tensile test (See Figures 6 and 15) which is primarily a function of the strength of the individual fibers show that the strength of the fiber tended to decrease only slightly with repulping.

Inasmuch as the external surface area--i.e., specific surface--of the stock remained essentially constant with repulping the amount of bonded area in the handsheets made therefrom should remain potentially the same. It may be seen by referring to the data tabulated in Table II and Figure 9, that the bonded area decreased only slightly with repulping, thus confirming for the most part the above projection.

Because of the nature of the repulping action, fiber length would be expected to decrease and the amount of fines increase. Since the specific surface and fiber classification data showed stocks without these effects, it may be concluded that some of the fines were lost in the processing steps. It is known (see page 49) that about one per cent of fiber by weight was unavoidably lost during the hand-sheet forming. Although relatively small on a weight basis, this fine fraction could have an appreciable effect on the specific surface of the stock had it been completely retained.

As mentioned earlier, because of the poor precision of the compressibility measurements, the specific volume data in Table IV shows no recognizable trend with repulping. However, effective specific volume, taken as a measure of the degree of swelling, should correlate with the degree of swelling measured by the centrifuge (Jayme) method. A plot of these data is shown in Figure 32 and it may be noted that the points scatter badly from a precise relationship. The centrifuge method for measuring swelling, although subject to a number of criticisms, shows a clearly defined relationship with repulping (see Figure 31). The lack of correlation is believed to be due to the variability introduced in the specific volume data as a result of the lack of precision in the compressibility measurement. This latter condition was prompted by the use of the very thin pads, which had to be used because of high filtration resistance on repulping. As would be expected, the swelling decreases rapidly during the early stages of repulping and then begins to level off. The decrease in fiber swelling, and the increase in pulp compressibility at the same value of specific surface is in good agreement with the observed

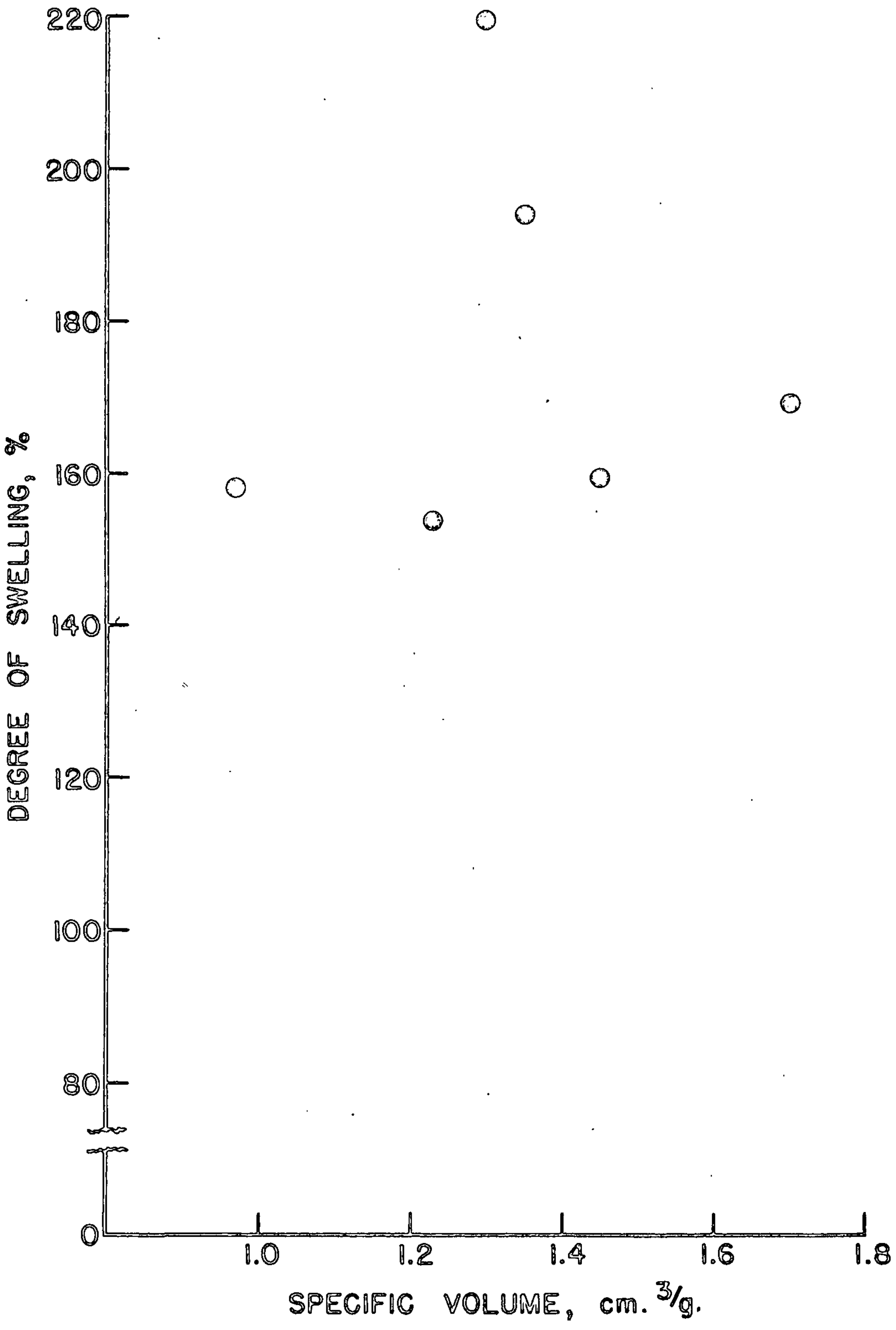

Fig. 32. Relationship of Effective Specific Volume and Degree of Swelling

behavior of filtration resistance and handsheet strength properties.

The strength of paper is a function of the strength of the individual fibers and the number and strength of the fiber-to-fiber bond. In the present data, the constancy of the specific surface values and the decrease in swelling, both of which are closely related to bonding, on repulping would lead one to expect a reduction in bonded area. However, since the amount of bonded area remained essentially constant, it may be postulated that the tendency for the bonded area to decrease with decrease in the degree of swelling was offset by some other attribute--e.g., fiber flexibility--which tended to increase the bonded area. However, as will be shown later, this latter increase in bonded area was not associated with a maintenance of bonding strength.

Inasmuch as the external surface area and the amount of bonded area in the handsheet remained essentially constant on repulping, any observed difference in strength properties would be expected to be attributable to either/or (a) decreased fiber strength, (b) increased fiber flexibility, or (c) a decrease in fiber-to-fiber bonding. The strength data shown in Tables II and III and illustrated in Figures 5 through 22, show a definite decrease in bursting strength, tensile strength, fold endurance, stretch characteristic, and VVP transverse bonding strength, and an increase in internal tear resistance with repulping. Also there is a tendency for zero span tensile (an approximation measurement of fiber strength) to decrease slightly with repulping.

In general, the effects level off with progressive repulping. The interplay of fiber and sheet properties indicate that the change in strength with repulping is primarily due to a decrease in the strength of the fiber bonds because the amount of bonded area remains relatively constant and the decrease in fiber strength is not appreciable with repulping under the condition used in this study. This view is also supported by the data obtained for the degree of swelling which is plotted against the various sheet properties in Figures 33—36.

The relationship between bursting strength and the degree of swelling is illustrated in Figure 33. It may be seen that the points align themselves in such a manner as to indicate a relatively good relationship between bursting strength and the degree of swelling. This is to be expected because bursting strength is primarily dependent on tensile and stretch, both of which decreased with repulping. The data plotted in Figure 34 show the relationship between the degree of swelling and tensile and stretch characteristics. Although there is some scatter, the relationship demonstrates that there appears to be a dependency of tensile and stretch on the degree of swelling. As in the case of bursting strength, this is to be expected because of the dependency of tensile and stretch on the number and strength of the fiber-to-fiber bonds and the relation of the degree of swelling to bonding.

The fold endurance plotted against degree of swelling is shown in Figure 35. On the basis of the present data, the correlation

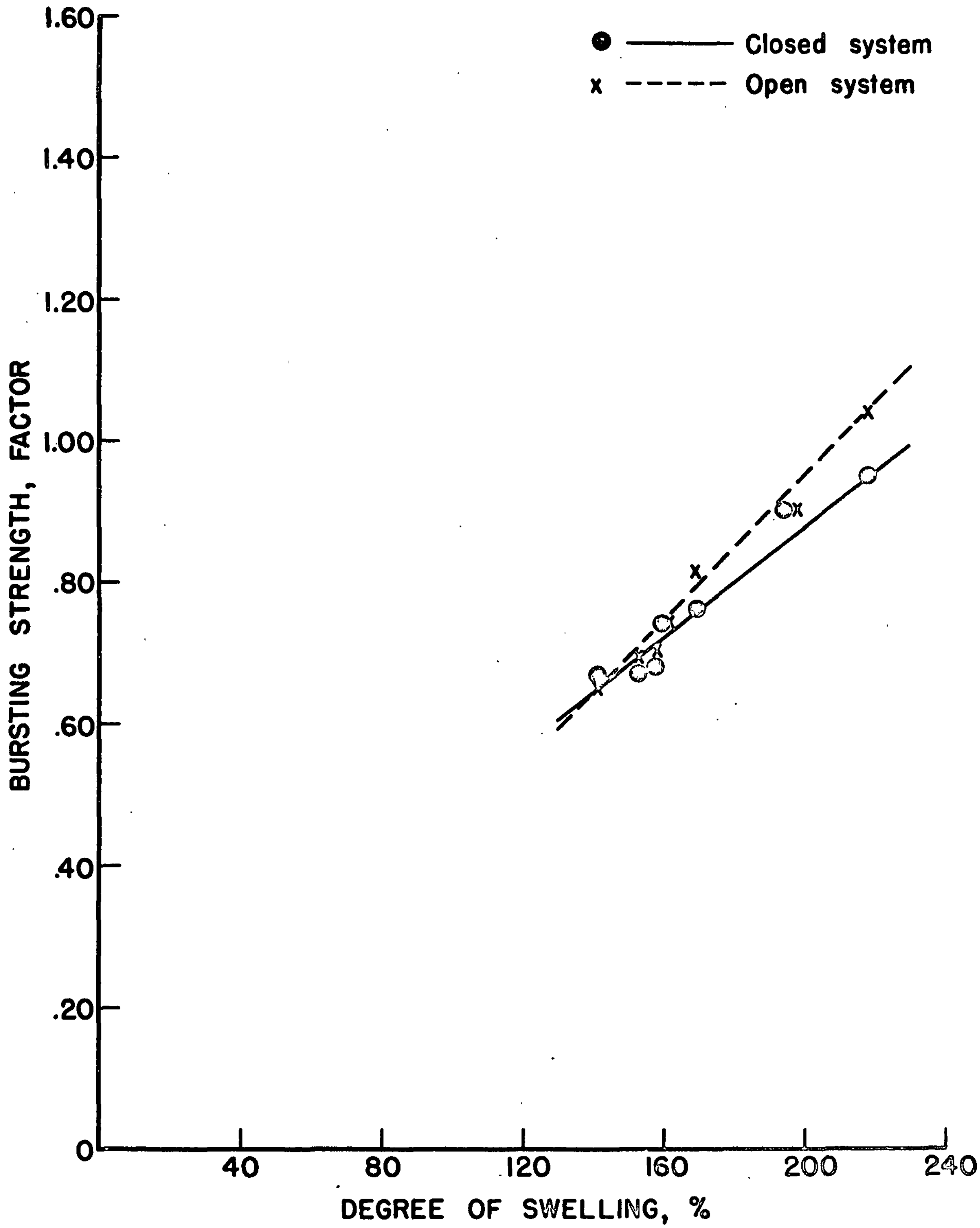

Fig. 33. Relationship Between Bursting Strength and the Degree of Swelling

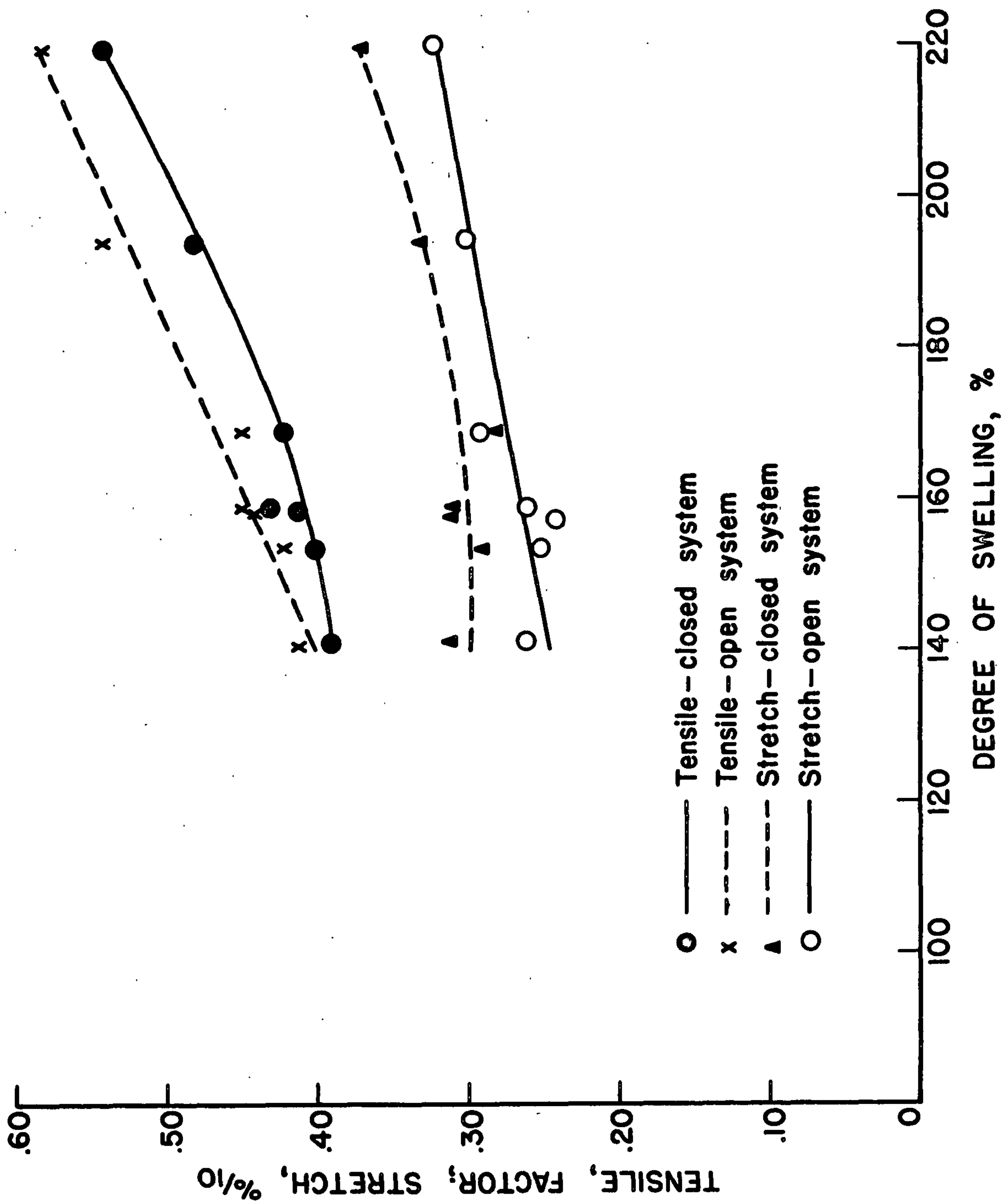

Fig. 34. Relationship Between Tensile Strength, Stretch, and the Degree of Swelling

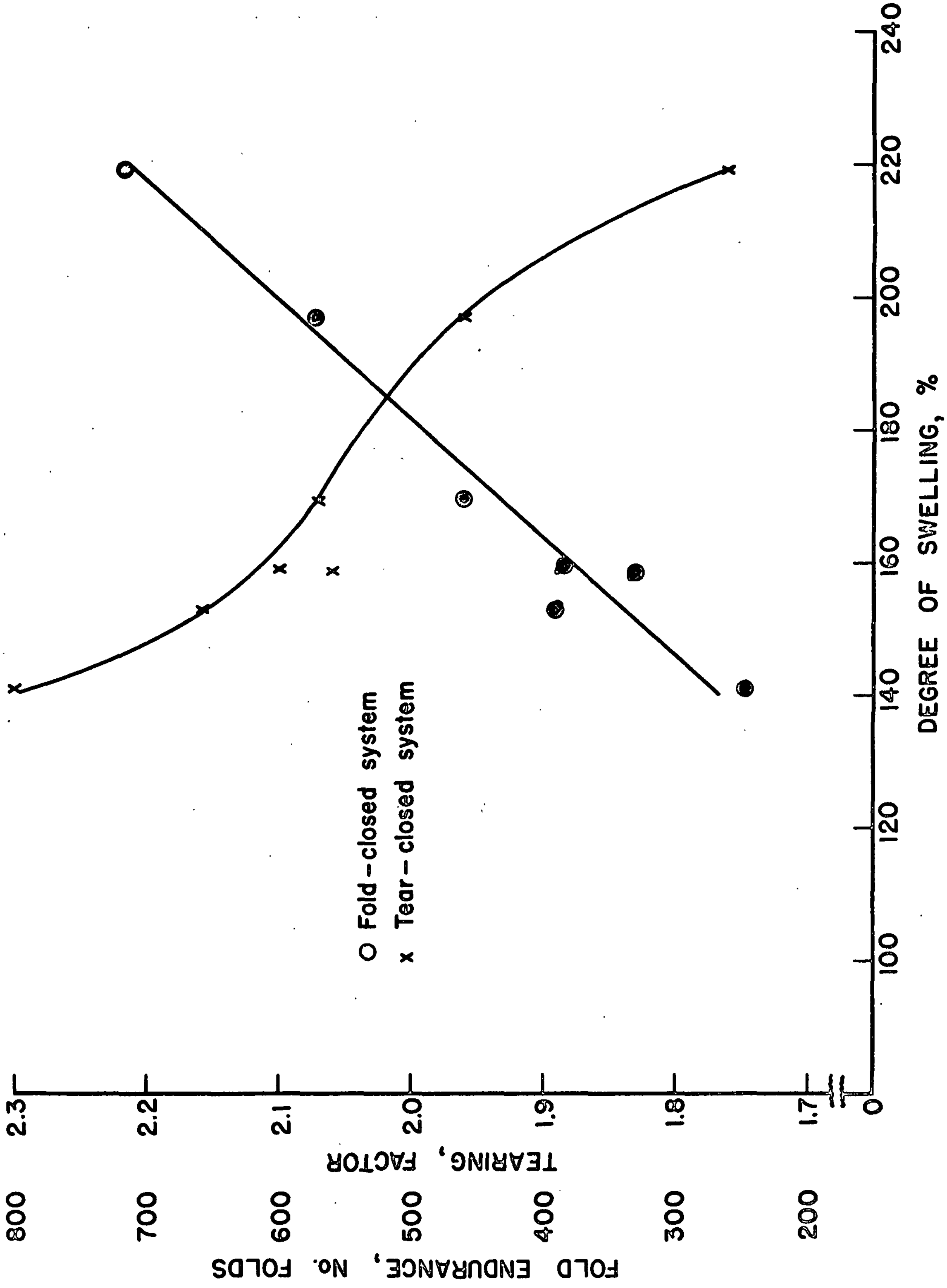

Fig. 35. Relationship Between Fold Endurance and Tearing Strength and the Degree of Swelling

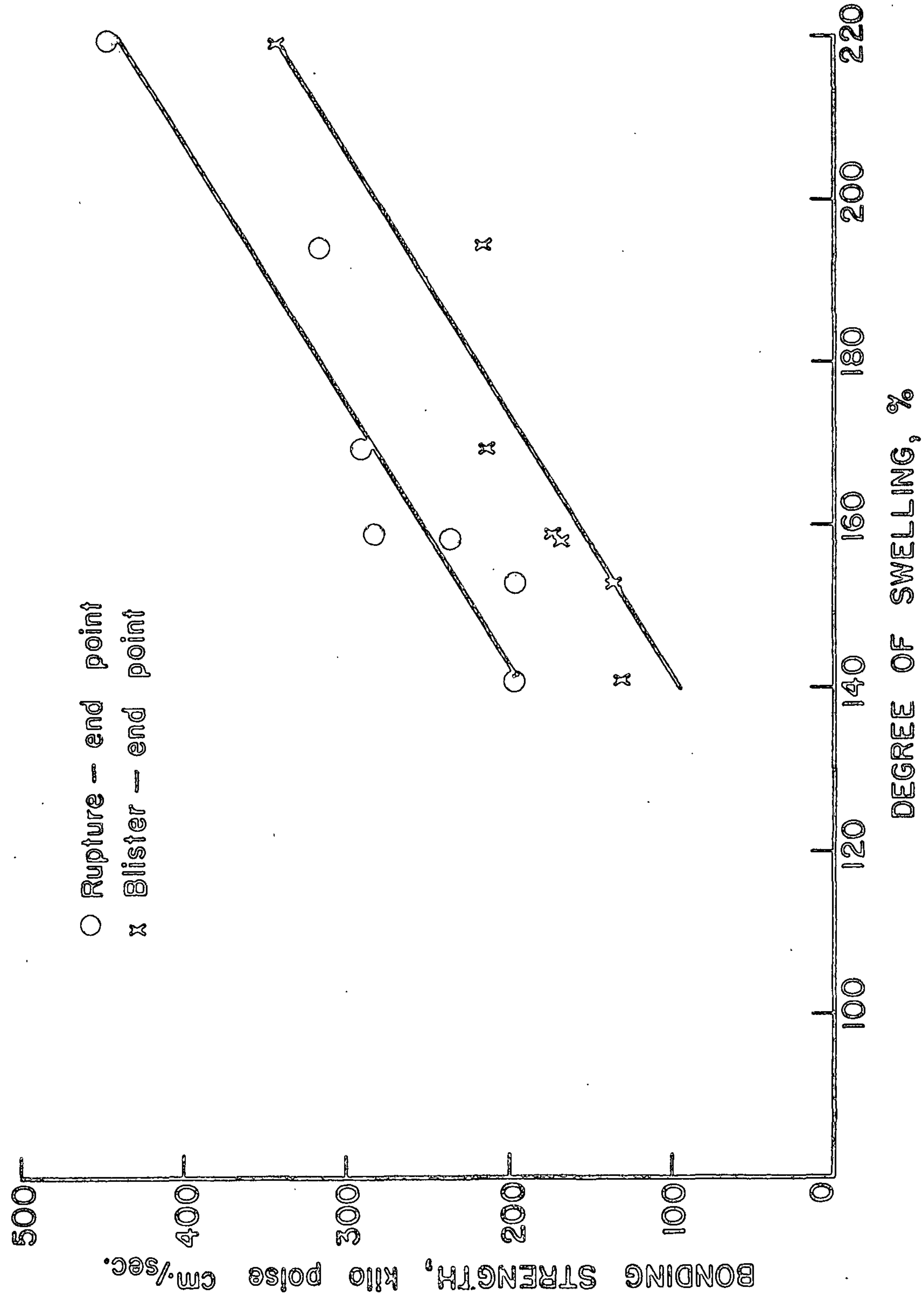

Fig. 36. Relationship Between VVP Bonding Strength and the Degree of Swelling

appears to be of a fairly high order indicating that fold endurance is directly related to the degree of swelling. Here again the relationship undoubtedly results from the relation of swelling and fold endurance to bonding strength. The tear strength <u>versus</u> degree of swelling is also shown in Figure 35. The results indicate that the tearing strength is inversely related to the degree of swelling and hence bonding. These results are in agreement with recognized behavior of paper properties since the fiber length remained relatively constant and the tear increased with lower swelling (lower bonding strength). Under the conditions of decreased swelling, the bonding was lower and consequently a higher tear value was obtained because a greater proportion of the fiber was being pulled out during the tear test. When the bonding was higher (higher degree of swelling), the tear values were lower.

The results of the VVP transverse bonding test when plotted against the degree of swelling show that these two properties are highly correlated, thus confirming the previous contention that bonding strength and swelling are closely related. The higher the degree of swelling, the greater the expected transverse bonding strength.

The change in strength properties as seen from the above comparisons all seem to be dependent for the most part on the number and strength of the bonds. Tests of bonded area indicated only a slight decrease on repulping; thus, the most logical conclusion to draw from the data is that the depreciation of strength is primarily caused by a decrease in the strength of the fiber-to-fiber bond. Undoubtedly, there is a certain depreciation of fiber strength taking place in repulping due to the action of the beater. However, the extent to which the fiber strength was lowered did not appear to be appreciable.

SUMMARY

It is common knowledge that in the manufacture of paper or paperboard from a waste paper furnish it has not been possible to attain the strength level characteristic of the fiber in the virgin state. Prompted by the desire on the part of the Jute Research Group to realize a greater share of the original strength in waste paper, a study was initiated to: (1) Study the effect of progressive repulping on the physical properties of the papermaking fibers and the strength of the sheets made therefrom; (2) develop basic information regarding the mechanism responsible for the change in strength properties; and (3) apply the basic information developed above in studying ways and means of obtaining a proportionately greater share of the original strength than has heretofore been attained with paper made with a waste paper furnish.

The present study is directed to objective 1: namely, to study the effect of progressive repulpings on the physical properties of the regenerated fibers and the strengths of the papers made therefrom. To this end a virgin unbleached kraft pulp was subjected to successive repulpings at a constant freeness level and the characteristics of the fibers and the resulting papers determined.

The procedure used in preparing the stocks was as follows: The virgin pulp was refined in a laboratory beater to a freeness of 615 ± 15 cc. S-R. and all the stock not used for stock tests was made

into standard weight handsheets. Except for a few sheets made with an open white water system and air dried, all the sheets were made with a closed white water system and dried on a steam-heated drier drum. The steam-dried sheets from the initial refinement, not used for test purposes, were used as the beater charge for the first repulping cycle. This procedure was used until a total of one initial refinement and six repulping cycles had been carried out. The procedure used in the six repulping cycles was identical to that employed in the initial refinement cycle.

The tests employed to characterize the properties of the fibers were freeness, pH, fiber length classification, filtration resistance and its components--specific surface, specific volume and compressibility--and the degree of swelling. The properties of the resulting papers were evaluated by means of caliper, basis weight, apparent density, bursting strength, tensile, zero-span tensile, stretch, internal tearing strength, porosity, Taber stiffness, VVP bonding strength, fold endurance and bonded area.

The results of the test on the handsheets prepared from the stocks subjected to progressive cycles of repulping indicate the following:

1. The bursting strength decreased rapidly with the number of times the stock was repulped. At the end of the sixth repulping, the bursting strength was 30 to 35% lower than was obtained on the first

refinement cycle, even though the stocks were refined to the same freeness level. In addition, it was observed that the most rapid rate of decrease in bursting strength was observed for the first and second repulping cycles.

2. The tensile strength of the handsheets made from stock subjected to progressive repulping decreased with the number of times the fibers were repulped. The decrease in tensile strength after six repulpings amounted to approximately 30%. As was noted for bursting strength, the majority of the loss in tensile occurred in the first and second repulpings.

3. The zero-span tensile, which is primarily a function of the intrinsic strength of the individual fibers did not experience any appreciable change in the first and second repulping cycles. The sheets made with the closed system and drum dried did not exhibit any appreciable change through the six repulping cycles. On the other hand, the sheets made with an open white water system and air dried exhibited a marked decrease in zero-span tensile in the 3 to 6 repulping cycles. Thus, it would appear that the significant changes noted above for bursting strength and tensile took place in the absence of any significant change in the structural strength of the individual fibers.

4. The stretch characteristics of the handsheets decreased with increasing repulpings. At the end of the sixth repulping cycle, the percentage stretch was approximately 15 to 16% lower. The most rapid depreciation of this property occurred in the first and second repulping cycle and thereafter tended to approach a constancy.

5. The apparent density, which is a function of caliper and basis weight, decreased with increasing number of times the fibers were repulped. At the end of the sixth cycle, the apparent density was 10 to 12% lower. Since the basis weight of the handsheets was held at a given level, the caliper increased with repulping. The increase in caliper would appear to be associated with a decrease in bonding. Normally, the greater the bonding, the more compact are the fibers and the lower the caliper per unit weight. The highest rate of depreciation of apparent density occurred in the first two repulping cycles.

6. The results of the tearing strength determinations show that this property increased with the number of times the fibers were repulped. The tearing strengths for the sixth repulping cycle was 25 to 30% higher. The most rapid increase in tearing strength occurred in the first and second repulping cycles.

7. The results of the fold endurance test show that progressive repulpings introduced a marked decrease in fold endurance of the handsheets, the depreciation of fold at the sixth repulping cycle being in the neighborhood of 50 to 60%.

8. The magnitudes of the Taber stiffness results indicate that there probably was no significant change.

9. The porosity results varied quite widely and there is no clearly defined trend. However, there is an indication that porosity decreased with repulping which would be what might be expected in view of the indications of a loss in bonding strength.

10. The results of the VVP bonding strength test show a very sharp decline in strength for the first and second repulping cycles and then a rather steady decline thereafter. Because of the nature of the test, the results indicate rather strongly that the bonding strength decreases sharply in the first and second repulping cycles.

11. The results of the measurement of bonded area indicate that there is only a relatively small decrease in bonded area with increased repulping. Further, the decrease in bonded area for the first and second repulping cycles was very slight. Since the bonded area decreased only slightly, the results suggest that the loss in bonding was the result of a decline in the strength of the bonds rather than a change in the number of bonds or bonded area.

The data obtained from the tests used to characterize the quality of the stock at successive repulping cycles indicate the following:

1. The freeness, which was used as a criterion of constant beating was held within a range of ± 15 cc. at a level of 615 cc. S.-R. The amount of beating to attain this freeness level decreased with each repulping.

2. The pH of the stock did not vary appreciably and may reflect only the variation in the pH of the water used in beating the stock.

3. The fiber classification data indicates that repulping under the conditions used in this study introduced only minor changes in the length of the fibers as determined by the Bauer-McNett classification test.

4. Filtration resistance data obtained by the constant flow method worked out by Ingmanson and Whitney show that the filtration resistance, which is a measure of drainage, increases approximately 30% on the first repulping. The increase in successive repulpings after the first (2 to 6) was very minor. Thus, the results show that the physical and chemical changes introduced in the fibers as a result of environmentaleffect of the initial refinement and subsequent drying decreased the ease of drainage by approximately 30%. Further, the results indicate that on the basis of this study the freeness level is not a good indication of the drainage characteristics inasmuch as the filtration resistance increased 30 to 35% without any significant change in freeness.

5. Analysis of the data for the components of filtration resistance--i.e., specific surface, specific volume and compressibility indicate that the specific surface was not altered significantly, the specific volume varied rather widely due to method difficulties, and the compressibility increased with progressive repulping. The latter behavior may be due to an increase in the flexibility of the fibers.

6. The results of the swelling measurements by the centrifuge method show that swelling decreased with progressive repulping cycles. The degree of swelling at the end of the sixth repulping cycle was approximately 35% lower than on the initial cycle. The highest rate of decrease in the degree of swelling occurred in the first and second repulping cycles.

As was pointed out in the body of this report, the strength of a sheet of paper is dependent on the intrinsic strength of the individual fibers and/or the fiber-to-fiber bonds. The latter are dependent on the number of bonds as well as the strength of the bonds. Thus, the failure of a sheet of paper results from (a) rupture of individual fibers, (b) a rupture of the fiber-to-fiber bonds, or (c) a combination of these. The strength of papermaking fibers is usually developed by subjecting them to mechanical attrition--beating or refining. The beating action may be considered as having a two-fold effect on the fiber. As pointed out earlier, the purpose of the beating action is to open up the structure of the fibers so that water can enter the amorphous cellulose portion of the fiber and swell it. In addition, the swelling is accompanied by a cutting, brushing, bruising, crushing and fibrillation of the fibers. The swelling appears to activate the surface of the fibers which, coupled with the fibrillation or unwinding of the fibrils, probably constitute the major action responsible for fiber-to-fiber bonding. On the other hand, the cutting, bruising, crushing and fibrillation or unwinding of fibrils all tend to reduce the intrinsic strength of the fiber.

An analysis of the change in sheet properties in terms of the properties of the fibers at corresponding levels of repulping make possible certain generalizations as to the cause of the strength changes.

1. The results of the zero-span tensile test indicate that there was little or no significant change in the intrinsic strength of the

individual fibers. This is particularly apropos to the first and second repulping cycles, which, it may be recalled, exhibited the greatest rate of change in strength properties. This conclusion is confirmed by the constancy of the specific surface measurements and also by the fact that the fiber-length distribution was not altered significantly on repulping by the beating technique employed in this study. The foregoing therefore suggests that the major depreciation in sheet strength on repulping is the result of a reduction in fiber-to-fiber bonding rather than a decrease in the intrinsic strength of the fibers.

2. Inasmuch as the external surface area--specific surface--of the stock and the amount of bonded area in the handsheets remained essentially constant on repulping, any observed change in strength properties of the sheet would be expected to be attributable to a decrease in the strength of the individual fiber-to-fiber bonds. This conclusion is confirmed by the data obtained for the degree of swelling and also by the bonding strength data. Since the reduction in VVP bonding strength was not accompanied by a reduction in bonded area, the reduction must have been the result of a progressive decrease in the strength of the individual fiber-to-fiber bonds. It is well known that bursting strength, tensile, stretch, fold endurance and VVP bonding strength are highly dependent on fiber-to-fiber bonding. In turn, the degree of swelling is directly related to the sheet bonding potentials. The tearing strength, on the other hand, is inversely related to the sheet bonding and, consequently, to the degree of swelling.

An analysis of the strength data in terms of the degree of swelling indicates that bursting strength, tensile strength, stretch, fold endurance, and VVP bonding strength appear to be directly related to the degree of swelling. The internal tearing strength, on the other hand, appears to be inversely related to the degree of swelling. Further, the greatest change in swelling (cycle 1 and 2) coincides with the greatest rate of change in the strength properties. All these behaviors point strongly to the conclusion that the depreciation of sheet strength is attributable to a loss in bonding as a result of repulping. Further, since the loss in bonding occurred in the absence of an appreciable change in bonded area, the change in strength properties would appear to result from a decrease in the strength of the fiber-to-fiber bonds and not a decrease in the number of bonds.

Inasmuch as the primary cause of strength depreciation on repulping appears to be a decrease in the strength of the fiber-to-fiber bonds, it may be postulated that, if a practical means of increasing the bonding--i.e., either the number of bonds or the strength of the individual bonds--were found, greater sheet strength would be realized in papers made from repulped waste paper. Further, the results of this study indicate that one of the most obvious approaches would be through the "medium" of swelling.

FUTURE WORK

The results which have been obtained in this study indicate that the primary cause of strength depreciation on repulping is attributable to a decrease in bonding strength. Analysis of the mechanisms and fiber properties responsible for bonding strength indicate that there are a number of ways in which the bonding strength may be improved. Among those which we feel should be investigated are:

1. Investigation of ways and means of increasing the degree of swelling and activation of the fiber surface. There is reason to believe that the environmental exposures to which the fibers are subjected in the initial cycle may in a sense "case harden" the fiber such that the surface activity is decreased. Among the variables which should be investigated in this phase are time, temperature, pressure, consistency and the action of chemicals.

2. Study the effect of chemical additives on bonding strength and the determination of the conditions most favorable to their bonding action.

3. Investigate the application of fibrous additives--stock "sweeteners."

4. Investigation of refining techniques which will provide the greatest reactivation of the surface with minimum deleterious effect on the intrinsic strength of the fiber and drainage characteristics.

THE INSTITUTE OF PAPER CHEMISTRY

R. C. McKee

R. C. McKee, Chief, Container Section

B. L. Sbertole

B. L. Sbertole, Technical Assistant

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